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Review

Plastic waste reprocessing for circular economy: A systematic scoping review of risks to occupational and public health from legacy substances and extrusion

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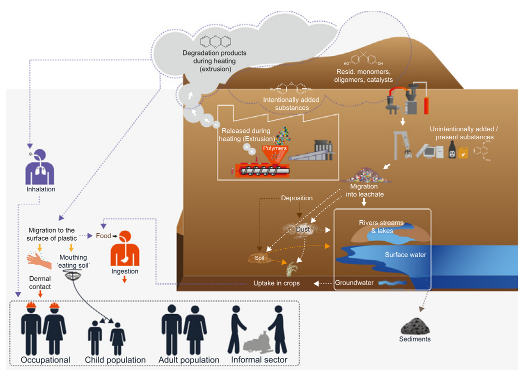
HIGHLIGHTS

- Increasing recycling of plastics may result in unintended consequences for health.
- A systematic scoping review focussed on legacy substances and extrusion activities
- Semi-quantitative risk assessment used to rank and prioritise risk scenarios
- Lack of safe systems of work in parts of Global South risk health of poorest
- Poor control of plastics reprocessing feedstock must be urgently addressed.

GRAPHICAL ABSTRACT



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ABSTRACT

The global plastics reprocessing sector is likely expand as the circular economy becomes more established and efforts to curb plastic pollution increase. Via a critical systematic scoping review (PRISMA-ScR), we focused on two critical challenges for occupational and public health that will require consideration along with this expansion: (1) Legacy contamination in secondary plastics, addressing the risk of materials and substances being inherited from the previous use and carried (circulated or transferred) through into new products when reprocessed material enters its subsequent use phase (recycled, secondary plastic); and, (2) Extrusion of secondary plastics during the final stage of conventional mechanical reprocessing. Based on selected literature, we semi-quantitatively assessed nine risk scenarios and ranked them according to the comparative magnitude of risk to human health. Our analysis highlights that despite stringent

Abbreviations: 8-OHdG, 8-hydroxy-2′-deoxyguanosie; ABS, acrylonitrile butadiene styrene; BaPeq, benzo(*a*)pyrene equivalent; BDE, brominated diphenyl ether; BFR, brominated flame retardants; Conc., concentration; DEHP, di(ethylhexyl) phthalate; ELV, end of life vehicle; EU, European Union; Geog., geographical context; Haz., hazard; HDPE, high density polyethylene; HIC, high income countries; IARL, indoor air reference levels; K-resin, SBC, styrene-butadiene copolymer; L, likelihood; LDPE, low density polyethylene; LIMIC, low income and middle income countries; LLDPE, linear low density polyethylene; Man'f, manufacturing; MDA, serum malondialdehyde; MEHHP, mono(2-ethylhexyl) phthalate; MEHP, monoethylhexyl phthalate; MEOHP, mono(2-ethyl-5-oxohexyl) phthalate; MiBP, mono-isobutyl phthalate; MnBP, mono-n-butyl phthalate; ND, Not detected; NSP, non-specified packaging; OR, odds ratio; PA, polyamide; PAH, polycyclic aromatic hydrocarbons; PBB, polybrominated biphenyl; PBDEs, polybrominated diphenyl ethers; PC, polycarbonate; PC-ABS, polycarbonate/acrylonitrile-butadiene-styrene; PE, polyethylene; PET, polyethylene terephthalate; PET(G), glycol-modified polyethylene terephthalate; phth., phthalates; POP, persistent organic pollutants; POP-BDE, persistent organic pollutant brominated diphenyl ethers; PP, polypropylene; PS, polystyrene; PTE, potentially toxic elements; Purch, purchased; PVC, polyvinyl chloride; pw, plastic waste; R, risk; RoHS, restrictions on hazardous substances; RPE, respiratory protective equipment; RPET, recycled polyethylene terephthalate; RQ, research question; S, severity; SD, standard deviation; SOD, serum superoxide dismutase; SVOC, semi volatile organic compounds; TBBPA, tetrabromobisphenol A; temp., temperature; TEQ, toxic equivalency; tonne, 1000 kg; TPAH, total polycyclic aromatic hydrocarbon; US, United States; USMR, uncertainty, strength of knowledge and methodological robustness; VOC, volatile organic compounds; VVOC, very volatile (gaseous) organic compounds; WEEE, waste electrical and electronic equipment; wt., weight.

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regulation, industrial diligence and enforcement, occasionally small amounts of potentially hazardous substances contained in waste plastics are able to pass through established safeguards and re-enter (cascade into) the next use phase (product cycle) after being recycled. Although many of these 'inherited' chemical substances are present at concentrations unlikely to pose a serious and imminent threat, their existence may indicate a wider or possible increase in pollution dispersion. Our assessment indicates that the highest risk results from exposure to these substances during extrusion by mechanical reprocessors in contexts where only passive ventilation, dilution and dispersion are used as control measures. Our work sets the basis to inform improved future risk management protocols for a non-polluting circular economy for plastics.

Contents

1. Introduction

Despite the great benefits engineered polymers (plastic materials and products) bring to society, plastics may also have substantial drawbacks, especially when they reach the end-of-life phase (Rochman et al., 2016; Burns and Boxall, 2018). Management of plastic waste is a pervasive, multifaceted and highly debated challenge of our times (Haward, 2018). Despite the recent calls and laws on limiting production of single use/ disposable plastic articles (Xanthos and Walker, 2017; da Costa et al., 2020), the consumption of plastics and hence plastic waste generation continues to follow an exponential growth curve (Geyer et al., 2017). A recent treaty signed by 175 countries of the UNEA (2022) is anticipated to catalyze a global effort to mitigate the negative impacts of plastic waste mismanagement (Velis, 2021; Silva Filho and Velis, 2022). However, current modeling suggests that plastic pollution will continue to increase, virtually unabated, under existing global policies (OECD, 2022). Three main narratives, partially intersecting, dominate the waste (or "after-use") debate:

- First, considerable attention is already paid to the fate and negative implications of plastic waste items when they are accidentally or purposely released into the environment, contributing to marine litter and wider plastic pollution (Bucci et al., 2020).
- Second, a recent imperative for a circular economy for plastics is gradually being established (Chen et al., 2021), exploring how resource recovery from waste can be extended to include re-use, remanufacturing, refurbishing, and product redesign along with waste avoidance and minimization (Korhonen et al., 2018).
- Third, non-negligible quantities, approximately 6.4 million tonnes in 2020 (OECD, 2022) of plastic waste (used/ secondary) are traded internationally; representing an integral component of the global circular economy for several decades (Velis, 2014). This trade is often characterized by unsorted mixed plastics exported from high income countries (HICs) to low- and middle-income countries (LIMICs), predominantly in South and South East Asia (Brooks et al., 2018). However, there are growing concerns that the residues from sorting and reprocessing these materials are being mismanaged in recipient countries (and may also be leaking into the aquatic environment) (Secretariat of the Basel Convention, 2019).

These three overlapping discussions about management of plastic waste occur against the backdrop of major failures/ challenges of waste and resource recovery systems across the Global South, alongside inefficiencies in the Global North. Specifically, in high-income countries, plastic waste is managed by being disposed of in landfill; recovered as fuel in energy-from-waste plants; or mechanically recycled (Tejaswini et al., 2022). Yet, this formal waste industry collectively has one of the highest occupational accident rates of all industrial sectors in many countries (Health and Safety Executive, 2018; Doherty, 2019). Disaggregation of safety reporting data for plastics reprocessing from the wider waste management category is problematic, but as plastics represent a substantial proportion (6.4–13 % wt.) of the composition of municipal solid waste (Kaza et al., 2018), we can surmise that risks to workers in the plastics reprocessing category may also be high.

In LIMICs the picture is more varied. Around 2 billion people receive no municipal solid waste collection service (Wilson et al., 2015) and have to self-manage, mainly by scattering on land, or more commonly by open burning (Velis and Cook, 2021), estimated by Lau et al. (2020) at 18 and 49 million tonnes (Mt) per annum respectively for the plastic fraction. Virtually all of the material collected for recycling in LIMICs is carried out by waste pickers (Velis et al., 2022); informal entrepreneurs who may number between 10 and 20 million (Wilson et al., 2015; Lau et al., 2020). Plastic sorting and reprocessing operations are often smaller, and in some cases poorly regulated, without any environmental or occupation and public health protection in place (Kosgeroglu et al., 2004).

Fundamentally, and historically, waste management arose from the imperative to protect human health (Velis et al., 2009; Velis and Mavropoulos, 2016); a goal largely achieved in the Global North as a result of investment and technological advances (Cook and Velis, 2020). Hence, there has been a shift in scientific research from quantifying and mitigating such risks towards the opportunities of resources recovery; and even further of a wider circular economy (Héry and Malenfer, 2020). However, as we have just indicated, waste related risks persist in parts of the world, and potentially affect everyone via globalized secondary supply chains (Cook and Velis, 2022), our shared atmosphere, and our delicate ecosystems (OECD, 2022).

The system through which plastic waste flows across society and the environment (Supplementary Information: Section S1, Fig. S1), is in many ways similar to other major constituents of solid waste. However, plastic waste includes many thousands of engineered plastics grades (polymer, filler and additive combinations) which exhibit persistent and fragmentary behaviour in the environment when mismanaged (Barnes et al., 2009; Teuten et al., 2009). Whereas the plastics system itself has been the subject of several global and national studies, for instance Geyer et al. (2017), Bai et al. (2018), and Lau et al. (2020), surprisingly or not, there have been few systematic efforts to quantify and compare the related risks between human health and plastic waste.

Several general reviews of solid waste management exist that summarize health and safety challenges (Giusti, 2009; Searl and Crawford, 2012; Ferronato et al., 2019). Although they include plastic waste as a component of the overall solid waste category, it is one of the least focused materials, being relatively benign in comparison to other hazards such as pathogen infection or road traffic related incidents. Two exceptions exist in the grey literature. Williams et al. (2019) focused on open burning, calculating a ballpark global mortality rate associated with improper management of all waste at between 400,000 and 1 million people per annum; inferring that plastic waste specifically may be responsible for a considerable proportion of these estimated deaths. Azoulay et al. (2019) made no attempt at inferring the magnitude of harm to human health from plastic waste, but highlighted and discussed a substantial list of potential hazards which may be associated with it, identifying pathways, but without determination of potential exposure.

One of the challenges for determining exposure to harmful substances from waste plastics is that public information on the composition, prevalence or hazardousness of the large array of plastic additives is limited (OECD, 2009; Groh et al., 2019; Wiesinger et al., 2021). For instance, Groh et al. (2019) identified >4000 substances potentially used in plastic packaging, of which 131 were assessed to be hazardous to human health (63) or the environment (68). Encompassing the whole plastics category, Wiesinger et al. (2021) investigated >10,000 substances, of which >2400 were highlighted as 'substances of concern' because of their hazardous, bioaccumulative or environmentally persistent properties. In Europe, the potential hazardousness of chemicals is assessed under the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) (European Union, 2006) and/or the Classification, Labelling and Packaging (CLP) regulations (European Union, 2008). However both Groh et al. (2019) and Wiesinger et al. (2021) found that approximately 60 % and 25 % respectively of substances they investigated have not undergone or registered any kind of hazard classification.

Many potentially hazardous substances in plastics (for example unreacted Bisphenol-A, or phthalate plasticizers) never make contact with any lifeform and remain within the material matrix throughout their lifecycle (Zeng et al., 2022). However, there have been justified concerns for several decades that certain harmful substances may leach from the surface of plastic objects and into the environment, plants, animals or humans (Golberg, 1963; Tice, 1998; Crompton, 2007). Hahladakis et al. (2018) investigated some of these pathways and described the fate and mode of transfer of plastic additives through the managed, mismanaged and unmanaged plastic waste systems. They highlighted that the plastics reprocessing pathway may be vulnerable to the presence of plastic additives but also so called 'non-intentionally added substances'. If present in the plastics reprocessing feedstock, these substances could be transferred into new plastic products, resulting in exposure to receptors in a subsequent use phase.

The presence of so many known, unknown, intentionally, and non-intentionally added substances represents a threat to the reprocessability of waste plastics and hence a barrier to implementing a circular economy for this complex group of materials (Cook et al., 2022). Therefore, for the first time, we bring some of the above concerns into focus by ways of systematically reviewing, consolidating and analyzing the available literature to provide an overview of the most significant human health risks that could be associated with plastic waste reprocessing. We group the information into key thematic areas dealing with (i) legacy substances present in plastic materials; and (ii) prevalent (manual/mechanical) reprocessing operations. Plastic marine litter and its effects upon animals, habitat, and humans are out of scope here. We also exclude the open burning of plastic waste, for which a separate review has been presented by Velis and Cook (2021). In addition, there were only two relevant papers revealed (Černá et al., 2017; Cioca et al., 2018) that addressed the sorting phase, and in each case, fell outside the scope of the remaining works; these were also not elaborated - we suggest that they belong in another in-depth study.

2. Methods

2.1. Systematic review

The present study is a component of a larger study that reviewed evidence for risks to health and safety from the wider topic of plastic waste mismanagement (Royal Academy of Engineering, 2021). This paper focuses on plastics extrusion and legacy substance contamination in secondary plastics, whereas another by Velis and Cook (2021) concentrates on the open, uncontrolled burning of plastic waste. The two reviews use the same initial pool of literature obtained through a systematic search, sharing some references across both as detailed in Section S.3.7. A brief summary of the methods is provided here, with more comprehensive details presented in Section S.3. The PRISMA-ScR checklist stipulated by Tricco et al. (2018) lists the specific requirements of the method and their location and can be found in the Supplementary Information: Section S2.

A systematic scoping review (Section S3.2) explored two research questions (RQ) following the PRISMA-ScR guidelines (Peters et al., 2020):

- RQ1: What evidence exists to indicate risk to public and occupational safety posed by plastic waste?
- RQ2: What are the comparative risks to public and occupational safety that arise from the management of plastic waste?

As recommended by Gusenbauer (2019), we searched Scopus, Web of Science and Google Scholar to improve the probability of capturing all literature (Section S.3.2.1). Boolean search terms were tested with one-at-a-time sensitivity analysis (Hamby, 1995; Xu et al., 2004) to ensure that they captured the maximum number of relevant papers (Section S.3.3). Titles were screened by a single reviewer according to pre-defined criteria (-Section S.3.5) and periodically checked by a second reviewer to ensure a consistent approach (Section S.3.6). Abstracts were screened by two reviewers. Further searches were carried out such as snowball and citation searching (Cooper et al., 2018). Online datasets and libraries were also searched, from organizations such as The World Bank (2020), International Labour Organization (2020), World Health Organization (2020), WIEGO (2020), Global Alliance of Waste Pickers (2020), Health and Safety Executive (2020b) in the UK.

Information sources were categorized by the type of waste management activity and these were further distilled into two overarching activity based "challenges": 1) Legacy contamination of secondary plastics; and 2) Extrusion of secondary plastics.

2.2. Uncertainty, strength of knowledge and methodological robustness

Where appropriate, information was qualitatively coded on a case-by-case basis according to uncertainty, strength of knowledge and methodological robustness (USMR); footnotes below each table provide details in each case. Data or information that fell inside the scope of the inclusion criteria were assumed to be adequate unless marked as: (i) inconsistent or ambiguous description of sampling and sample processing; (ii) issues of comparability with data reported by different authors; and, (iii) comparability affected by age of study.

2.3. Conceptual diagrams

Identified risks and/or hazards were coded according to the type of hazard, risk, and the pathway through which the hazard may reach a receptor and the receptors themselves. Such evidence documentation and theorizing, allowed the creation of generalised hazard-pathway-receptor diagrams, visualizing risk for "Challenge 1" and "Challenge 2". These are shown in each of the Sections 3 and 4, whilst a combined version, although it is a result, is shown here in the method to assist understanding (Fig. 1).

2.4. Risk based approach

An approach to summarising and ranking risk to human health, a semi-quantitative method (Section S.3.3) adapted from World Health Organization (2012), Hunter et al. (2003), Kaya et al. (2018) and Burns et al. (2019), was undertaken to indicate and rank the relative harm caused by different activities (Table S4). As suggested by Kaya et al. (2018), this method was not intended to quantify risk associated with the identified hazard-pathway-receptor combinations or inform decisions directly. Rather it was intended to support decision-making and indicate where efforts for intervention or further research might be directed.

For each Challenge, hazards and risks identified in the literature were grouped into hazard-pathway-receptor combinations alongside a qualitative assessment of the vulnerability of each receptor. Each combination was assigned a likelihood and severity score according to criteria detailed in Tables S5 and S6. The product of the likelihood and severity resulted in a color coded risk score assigned using Tables S7 and S8. The results of this scoring / ranking process are reported in Section 5 and also aggregated and ranked according to the risk scores in Table S9, Section S5.

3. Results: safety challenge 1 - legacy contamination in secondary plastics

When plastics are recycled, substances from their previous use are carried through into "secondary materials" (pellets or flakes), and subsequently into new plastic products that may have a different intended use to the original (Schyns and Shaver, 2020). Plastic additives that are transferred through the circular economy material chain in this way are referred

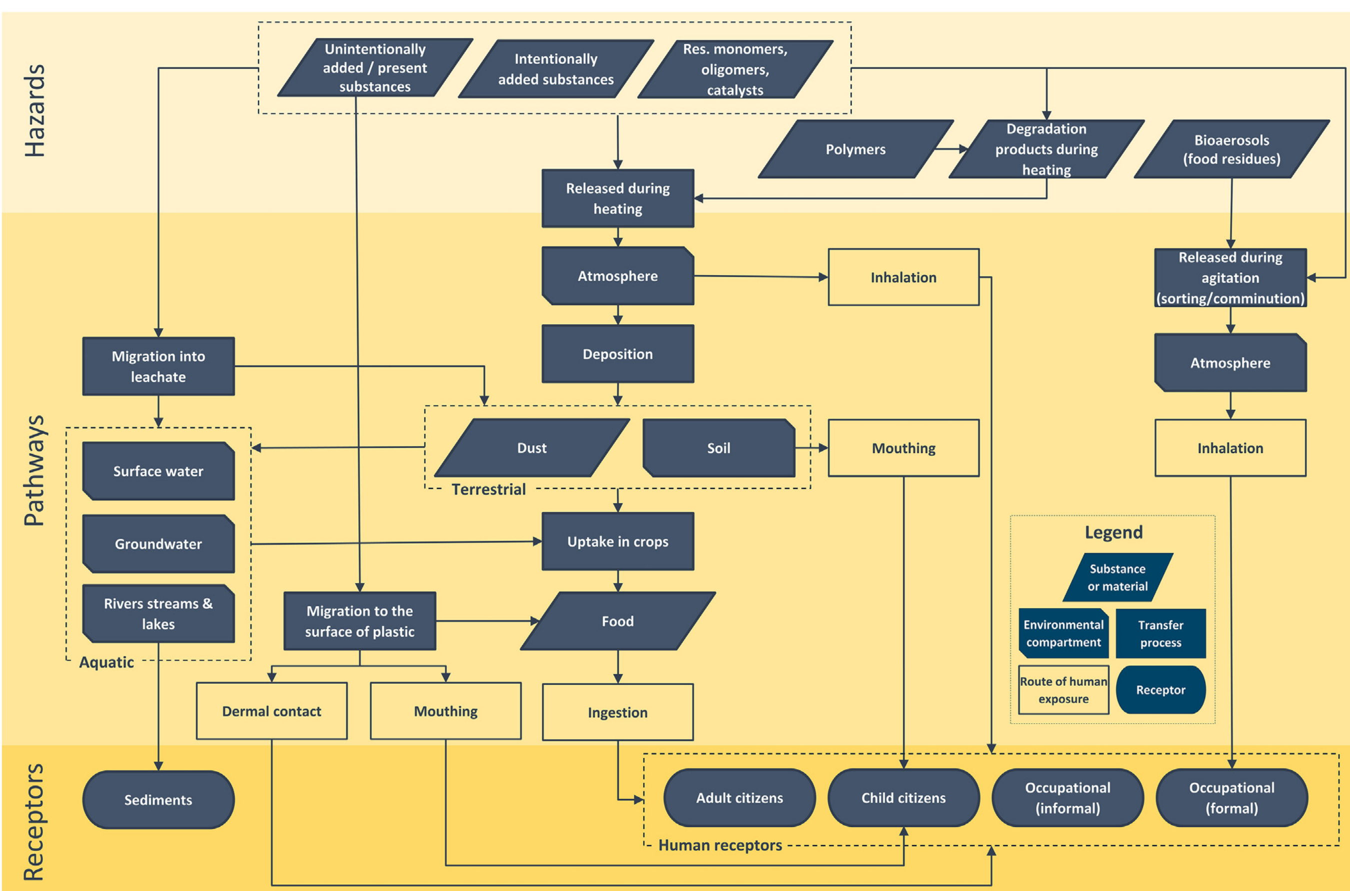

Fig. 1. Summary of the main hazards associated with the reprocessing of plastic waste and the pathways through which they may result in exposure to receptors. Based on scenario hazard-pathway-receptor combinations; method discussed in Section 2.3; results presented in Sections S.5 and 5.

to as "legacy additives" (Wagner and Schlummer, 2020) or "non-intentionally added substances" (Pack et al., 2021; Wiesinger et al., 2021); however, here we use the term "legacy substances", to encompass a wider range of substances as follows:

- Substances intentionally added to primary polymers to modify their characteristics such as bulking agents, impact modifiers, flame retardants (Hahladakis et al., 2018);
- Residual substances from primary plastic production such as unreacted monomers, catalysts and oligomers (Geueke, 2018);
- Residues of materials that have become attached (adhered to, adsorbed) to the surface of plastics or which have been absorbed into the space between polymer chains (hereafter unintentionally added substances), which can be categorized as:

  - ○ Residues that have arisen during the use phase (e.g. cooking oil which has sorbed onto the surface of a polyethylene terephthalate (PET) bottle; food which has become attached to the surface of an item of food packaging; garden pesticides which have been absorbed into a high density polyethylene (HDPE) milk bottle that has been repurposed) (Roosen et al., 2020); and
  - ○ Residues that have arisen during the end-of-life (after-use) phase (for example, engine oil which has become attached to the surface of an item of food packaging after being deposited in household recycling; battery acid that has leaked onto the surface of plastics during e-waste comminution) (Huysveld et al., 2019).

Several pathways exist through which people may be exposed to these substances during the second use phase illustrated in a conceptual diagram (Figs. 2 and S4). The arrows represent the pathways through which potentially hazardous substances may move and come into contact with people and the route of exposure.

In many cases, these legacy substances exhibit benign characteristics and/or occur in very low quantities; posing little risk to human health (Wagner and Schlummer, 2020). Even when they occur in larger quantities or are potentially hazardous, if they are bound to the polymer or have low migration potential, they may never transfer into surrounding media such as food or human skin (Pack et al., 2021).

The evidence for the occurrence of legacy substances reviewed here is grouped into sub-sections according to the following four groups of potentially hazardous compounds:

- Brominated flame retardants (BFR)
- Phthalates
- Potentially toxic elements (PTEs)
- Volatile organic compounds (VOCs)

In addition, each section reviews the comparatively small number of research outputs that have modeled risk to human health from these substances.

3.1. Brominated flame retardants

This group of substances, bromophenols, hexabromocyclododecane, polybrominated diphenyl ethers (PBDEs) (hereafter BFRs), have been added to plastics to inhibit combustion chemistry (Alaee et al., 2003) in automotive, electrical, aeronautical and furnishing applications since the 1950s (Covaci et al., 2011; Wagner et al., 2019; Sharkey et al., 2020). They are not used in food packaging as there is little need for flame retardant properties. Certain BFRs, particularly polybrominated diphenyl ethers (PBDEs), represent a risk to human health as they can disrupt the endocrine system and cause developmental neurotoxicity (Hong-Gang et al., 2016; McGrath et al., 2017).

Historically three major formulas of PBDEs have been in use: Penta-brominated diphenyl ether (BDE), Octa-BDE and Deca-BDE, between them comprising 209 congeners. All are classified as persistent organic

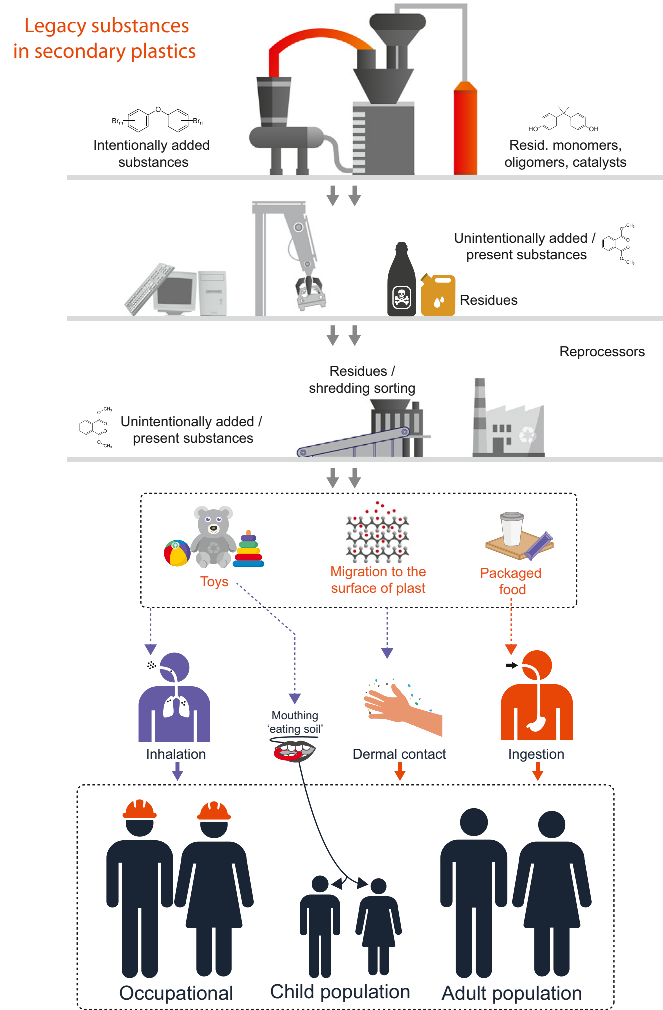

Fig. 2. Hazard exposure conceptual model (hazard–pathway–receptor) associated with legacy substances in plastic waste.

pollutants by the Stockholm Convention (Tang et al., 2014; UN Environment, 2017). Production of Penta-BDE and Octa-BDE is banned in the United States (US) (Venkatesan and Halden, 2014) and Europe (Alaee et al., 2003), however it still continues elsewhere, including in China (Tang et al., 2014; Yu et al., 2016); meaning the two congener groups are still at risk of entering the environment. Many products containing PBDEs remain in use today and are likely to continue to do so for many years to come (Covaci et al., 2011). In Europe, several legal instruments restrict the content of BFRs in secondary plastics as follows:

- Directive 2011/65 (European Union, 2011b); hereafter the Restrictions on Hazardous Substances (RoHS) Directive, restrict content of BFRs to 1000 $\mu g.g^{-1}$ (0.1 % wt.) plastic in electrical and electronic items;
- Directive 2012/19 (European Union, 2012); hereafter the Waste Electrical and Electronic Equipment (WEEE) Directive, states that substances, mixtures and components containing BFRs must be removed from separately collected e-waste;
- Directive 2009/48 (European Union, 2009); hereafter the Toy Safety Directive, states that substances that are mutagenic, toxic for reproduction or carcinogenic should not be used in toys;
- European Commission Regulation 2016/460 (European Union, 2016); hereafter the Persistent Organic Pollutant (POP) Regulations, states that materials with a concentration of BFRs exceeding 1000 $\mu g.g^{-1}$ cannot be recycled until their PBDE and hexabromocyclododecane content has been destroyed or irreversibly transformed. At the time of writing, a

Table 1
Brominated flame retardants (BFRs) identified in secondary plastics. Where multiple low concentrations were reported, only the highest concentrations are shown.

Ref.	Context	Samples	Substance	Mean or range conc. (μg.g^{-1}) plastic	Key findings	USMR[#]
			ΣPBDE	53	Median conc. in hard toys >	
			ΣDBDPE	5.54	others toys all <WEEE and	
			ΣBTBPE	0.101	RoHS Directives (1000 μg.	
					g^{-1}) Except single sample	
Chen et al. (2009)	CHN[a]	Assorted toys[b]	ΣPBBs	0.0279	5344 μg.g^{-1}	
		Rubik's cube		328.1		
		Toy gun		4352.7*†		
		Spring car		1303.8*†		
		Spring car		944.4†		
		Car launcher		9225.8*†		
		Miniature car		284.3		
		Miniature car		1279.8*†		
		Spring gun		210.5		
		Thermal cup		778.8†		
		Thermal cup		775.2†		
		Radio back panel	decaBDE	5118.8*†	BFRs found most frequently	
			ΣdecaBDE	~200-10,000	in toys	
		Rubik's cube		386.8	$^{1}/_{3}$ food contact items	
		Toy gun		661.3†	contained Br	
		Spring car		774.4†	61 % contained Br	
		Spring car		278.1	45 % contained ΣdecaBDE	
		Car launcher		7747.1*†	>1000 μg.g^{-1}	
		Miniature car		927.2	Food contact articles sold on	
		Miniature car		208.4	European market are not	
		Spring gun		513.9†	produced exclusively with	
		Thermal cup		442.8	food-grade polymers, con-	
		Thermal cup		471.3	travening Regulation (EC)	
	Purch: ITA, CZE,	Radio back panel	TBBPA	<LOD	no. 202/2014	
	DEU;		ΣTBBP-A	~200–8000	Many conc. >1000 μg.g^{-1}	
Guzzonato et al.			ΣBr	4–17,000	contravening WEEE & RoHS	
(2017)	Man'f: CHN TUR[c]		ΣBTBPE & DBDPE	Trace	Directives	
		European ELV parts		0.2		
		US/Asian ELV parts		0.3–25,000*†		
		WEEE items		0.5–800†	Conc.'s indicate some legacy	
		Shredded car plastic		0.1–11	contamination of secondary	
		Shredded car & WEEE plastic (mix)		1–280	waste stream but at low	
		Shredded WEEE plastic		2–330	levels	
		Recycled plastic pellets		0.7–67	Upper range limit of	
		Insulation/carpet padding		0.001–0.04	BDE209 (a candidate POP)	
		Office & kitchen products		0.005	in toys cause for concern	
Leslie et al. (2016)	NLD	Plastic toys	ΣPOP-BDE	0.01–33	<0.06–800 μg.g^{-1}	
		Wash basin, litter basket, mat, plastic stool,				
		mop, kettle, PPR pipe, PE pipe, PVC pipe,		5.98	Very low content in all	
Lyu et al. (2015)	CHN, Beijing	slippers, luggage & folder	ΣPBDE	(0.45–21.30)	samples	A
			ΣPBDE	61.9		
			Σ		Results indicate	
		PVC wastes	hexabromocyclododecane	18.7	contamination from legacy	
			ΣPBDE	388.0	materials as BFRs not	
Hong-Gang et al.			Σ		believed to be widely used	
(2016)	CHN	PS wastes	hexabromocyclododecane	20.8	in PS and PVC	
		PS (residual packaging waste)		4.4		
		PP (residual non-packaging waste)		3.0		B
	DNK	NSP (residual non-packaging waste)	TBBPA	2.2		B
			Dibutyl phthalate s	8		
	CHN, DNK, DEU,		2,4,6-TBP	340		
	NLD	ABS (recycled)	TBBPA	26,000*†		
				0.5		
		PS (residual packaging waste)		5.1		
		Expanded polystyrene (residual packaging				
	DNK	waste)		330		
	CHN, DNK, DEU,	PS (virgin)		0.01		
	NLD	PS (recycled)		0.76		
		PET (non-packaging waste)		1.3		B
		NSP (non-packaging waste)		3.2		B
				0.27		
		NSP (packaging waste)	Σ	0.63		
	DNK	Foil laminated (packaging waste)	hexabromocyclododecane	0.19		
		PP (waste packaging)		(4)	The presence of BFR in	
	DNK	PET (waste packaging)		(1)	multiple samples indicates use	
		HDPE (virgin)		(2)	of secondary plastics in	
		LLDPE (virgin)	PBDEs (presence only -	(1)	applications which pose a risk	
Pivnenko et al.	CHN, DNK, DEU,	PP (virgin)	number of congeners	(3)	to human health such as food	
(2017)	NLD	PS (virgin)	detected in brackets)	(5)	contact materials and toys	

Table 1 (*continued*)

Ref.	Context	Samples	Substance	Mean or range conc. (μg.g^{-1}) plastic	Key findings	USMR[#]
		PET(G) (recycled)		(2)		
		PET (recycled)		(2)		
		HDPE (recycled)		(2)		
		LDPE (recycled)		(3)		
		LLDPE (recycled)		(1)		
		PP (recycled)		(2)		

[*] Conc. > RoHS and POP Directive (European Union, 2019) threshold of 1000 μg.g^{-1}.
[†] Conc. > proposed amendment to POP Directive (European Union, 2019) threshold of. 500 μg.g^{-1}.
[a] (South) Guangzhou City.
[b] Toys: Racing cars, vehicles, weapons, action figures and hand-held video game consoles (n = 30); foam toys (for example, mats, puzzles, swords) (n = 18); rubber/soft plastic toys (for example, Barbie dolls, teethers) (n = 15); textile and stuffed toys (for example, animals, dolls, Christmas toys) (n = 6).
[c] toys were purchased in Italy and Czech Republic and manufactured in China and Turkey.
[#] Uncertainty, strength of knowledge and methodological robustness (USMR) assessed qualitatively. It is assumed that there are no significant concerns unless marked as: A = sample size not available. Abstract in English but paper in Chinese and inaccessible at time of writing (details presented from comprehensive abstract); B = non-packaging samples not specifically attributed to intended use, which could indicate that presence of BFRs is not unexpected. Abbreviations: Linear low density polyethylene (LLDPE); low density polyethylene (LDPE); high density polyethylene (HDPE); polypropylene (PP); polystyrene (PS); polyethylene terephthalate (PET); non-specified packaging (NSP);, polybrominated diphenyl ethers (PBDEs); tetrabromobisphenol A (TBBPA); persistent organic pollutant brominated diphenyl ethers (POP-BDE); polybrominated biphenyl (PBB); glycol-modified polyethylene terephthalate (PET(G)); manufacturing (Man'f); purchase (Purch); end of life vehicle (ELV); waste electrical and electronic equipment (WEEE); acrylonitrile butadiene styrene (ABS).

proposal for an amendment (European Union, 2019) seeks to reduce this threshold to 500 μg.g^{-1};

- European Commission Regulation 10/2011 (European Union, 2011a); hereafter the Food Contact Regulations, prescribes migration limits for BFRs into foodstuffs or food-simulation solutions.

Whereas the RoHS thresholds do not indicate hazard exposure or risk, they provide a tangible benchmark from which to contextualize the identified concentrations of BFRs in plastics. In industry, prevention of BFRs arising in secondary plastics is controlled by risk assessments which combine traceability of source material with visual observations of incoming materials, supported by laboratory testing (Houston, personal communication, 27 November 2019). However, despite the stringent regulatory framework and industry support in Europe, BFRs (and many other potentially hazardous substances) have been found in new plastic products from which they are meant to be excluded (Table 1). Whereas many of the samples analysed in the six studies did not contain high concentrations of BFRs, there were 15 that exceeded the limit of 500 μg.g^{-1} of plastic proposed in the amendment to the POP Directive (European Union, 2019) of which eight exceeded the RoHS Directive threshold of 1000 μg.g^{-1} of plastic.

Of these, Leslie et al. (2016) having identified BFRs in end of life vehicle (ELV) car parts is perhaps the least concerning (Table 1). The source of the parts was not identified and therefore the BFR content may have been added legitimately during an era when BFRs were not prohibited. Conversely, the sample of recycled acrylonitrile butadiene styrene (ABS) analysed by Pivnenko et al. (2017) (26,000 μg.g^{-1}) demonstrates an unbroken chain through which the presence of substances such as BFRs have persisted in materials as a consequence of recycling. As with several studies reviewed here, the source of the sample was ambiguous, which makes it hard to benchmark the socio-geographical or regulatory context. Moreover, the intended future use of the recycled ABS was not stated, which means that its potential to cause harm cannot be ascertained. For instance, if the recycled ABS was destined for the production of children's toys, the presence of such a high BFR content would represent a risk to children who enjoy chewing pieces (Groot et al., 1998; Scientific Committee on Health and Environmental Risks, 2016). However, if the intended use was as an internal electronic component, it would be unlikely to result in significant exposure to individuals as BFRs are not generally highly volatile and people would be unlikely to handle internal parts with high frequency during the use phase.

The identification of BFRs by Guzzonato et al. (2017) in a wide range of children's toys is concerning. In particular, five samples exceeded the RoHS limit, in one case by a factor of nine, and three exceeded the POP Directive limit (European Union, 2019). While all the toys purchased in Europe were manufactured in The People's Republic of China or Turkey, and Guzzonato et al. (2017) did not state whether they had been certified for sale in Europe. Nonetheless, their presence in such high quantities highlights a weakness in European systems to protect people from exposure to BFRs in imported plastic products that contain recycled material. Furthermore, Leslie et al. (2016) combined the concentration data presented in Table 1 with data from interviews with stakeholders in the Dutch waste management sector to estimate that 22 % (wt.) "POP-BDEs" from waste electrical and electronic equipment (WEEE) plastics and 14 % (wt.) POP-BDEs from ELV plastics are recycled into new products in The Netherlands. Though the assumptions made by Leslie et al. (2016) are strongly driven by the opinions of stakeholders, even small quantities of BFRs being recirculated in this way should be a cause for further investigation to ascertain the scale of the potential transgression.

Lyu et al. (2015) and Chen et al. (2009) identified concentrations of BFRs in plastic products in China and used them to model exposure to humans and risk to health (Table S13). Both studies reported the main exposure pathway for BFRs to be through inhalation. In each case, the risk was considered very low and a very small proportion of exposure in the context of other sources of BFRs that mainly result in exposure through being ingested in food.

3.2. Phthalates

Phthalates are a group of man-made substances used increasingly in a variety of industrial applications, but primarily in plastics (80 % in polyvinyl chloride - PVC and cellulose polymers) where they are added to increase flexibility (Benjamin et al., 2017). Phthalates are not chemically linked to, but occupy the mesh space between polymer chains in plastics (Yang et al., 2019), and are therefore sensitive to changes in the surrounding environment such as pH, temperature (Annamalai and Namasivayam, 2017), and pressure (Zhang and Chen, 2014), which can cause them to migrate to the surface (Stanley et al., 2003). Once outside the plastic, phthalates may be absorbed into human skin; ingested directly; volatilized and inhaled; transported into soil; food; and potentially the entire biota (Benjamin et al., 2017). The lipophilicity of phthalates, means that they are easily absorbed into the bloodstream or other human fluids where they are transformed into metabolites, which can disrupt signaling in the endocrine system (Tian et al., 2018). In animal studies, this disruption has been shown to be carcinogenic with potentially irreversible effects (Simoneit et al., 2005); have the potential to disrupt metabolism (Petrovičová et al., 2016); and may affect the status of thyroid hormones (Wang and Wang, 2018). Human studies are limited and inconclusive and there have been criticisms of some animal studies as they tend to involve exposing subjects to much higher doses than humans would experience in their ambient environment and often only of a single phthalate species (Swan, 2008).

Phthalates are not deliberately used in food packaging or toys in the European Union (EU) or US and rarely in LIMICs except possibly in cases where flexible PVC is still used (Hahladakis et al., 2018). But the potential harm phthalates may cause to human health has elicited anxiety and confusion among some people in society over the extent to which plastics contain them and the level of exposure which people may be subjected to (Entine, 2011; Carter, 2012; Putrich, 2015). The presence of phthalates is near ubiquitous throughout the environment (Gao and Wen, 2016), and they have been found in the bodies of 98 % of adults in the US for instance (Zota Ami et al., 2014).

One area of concern is the occurrence of phthalates in recycled plastics as a result of contamination of reprocessor feedstock, indicated by two studies identified in this review (Table S14). In both studies (Simoneit et al., 2005; Pivnenko et al., 2016), phthalates were identified in materials or products where they are not added intentionally, namely non-PVC and non-cellulosic plastics. Contextualizing phthalate content by mass is not necessarily the most informative metric, because concentration alone does not indicate migration potential. EU legislation does not provide content threshold; however, in the US, the US Code (2008) and Consumer Product Safety Commission (2017) set a limit for content in toys and related articles of 1000 $\mu g.g^{-1}$. Both Simoneit et al. (2005) and Pivnenko et al. (2016) identified concentrations of phthalates in several examples that exceed this limit. However, the highest concentrations occurred in samples which contained unknown polymers, meaning it is possible that the samples contained phthalate plasticized PVC. In a specific example, the "roadside trash" sample analysed by Simoneit et al. (2005) contained PVC of unknown origin and it is hence unsurprising that 2164.7 $\mu g.g^{-1}$ (0.21 % wt.) was identified, given that plasticized PVC may contain between 10 and 70 % by weight (wt.) of intentionally added plasticizer.

Nonetheless, the presence of phthalates in such a wide range of samples (Table S14), however small, is an indication that phthalates are being transferred through the value chain from materials such as PVC where they have been intentionally added, into products such as PET packaging where they may pose a risk to human health in larger quantities (Pivnenko et al., 2016). In another study, Keresztes et al. (2013) measured concentrations of phthalates in water sold in PET bottles in Hungary, finding very small quantities of phthalates in all samples (data not shown). Although the study concluded that the quantities were no threat to human health, they indicate the presence of phthalates: either in the bottles themselves, the lids, or possibly from water that has been contaminated prior to or during storage at bottling plants (Leivadara et al., 2008; Liu et al., 2008).

Whereas the assessment of harm to human health carried out by Keresztes et al. (2013) was based on bottled water consumption, children were not considered. To address this gap in understanding, Lee et al. (2014) used the concentrations observed by Keresztes et al. (2013) to model human phthalate consumption in Denmark and in particular, two-year-old children, finding that paper and PET food packaging could be responsible for 18 % of their childhood exposure (Table S15). Moreover, the study estimated that 2–12 % wt. of all phthalates placed on the market may re-enter the European product cycle as a consequence of recycling both paper and plastic packaging.

While the studies summarised in Tables S14 and S15 indicate the need for further investigation into legacy phthalates, none specifically indicate concentrations of phthalates in food packaging or toys, the product groups most likely to result in human exposure. Only the study by Lee et al. (2014) indicated that contamination of PET might be a source, but this study is driven by concentrations identified in a single study (Keresztes et al., 2013), which may not be representative of packaging on the European market.

3.3. Potentially toxic elements

Some elements represent a potential hazard to human health due to their toxicity at relatively low concentrations. For instance, lead, chromium (VI), nickel and cadmium are all potentially carcinogenic and can inhibit growth in humans (Whitt et al., 2012). Cadmium can damage kidneys and lead to skeletal damage; lead can cause impairments to cognitive ability and reduced mental capacity in children; and antimony can cause skin, eye and lung irritation at relatively low concentrations. These elements are commonly described as "heavy metals", however this term is non-specific, and therefore the present study will follow the recommendation of Pourret and Hursthouse (2019) and describe them hereafter as potentially toxic elements (PTEs).

In the EU, the Food Contact Regulations (European Union, 2011a) set maximum migration limits for selected elements from plastic food contact packaging. For metal concentration, the RoHS Directive (European Union, 2011b) in Europe and the California Toxics in Packaging Prevention Act (2005) provide maximum thresholds for metal content in electrical equipment and food packaging respectively (Table S10).

PTEs are added intentionally to plastics as: anti-slip agents (Hahladakis et al., 2018); catalysts (Office of the Report on Carcinogens, 2018); flame retardant enhancers (Dimitrakakis et al., 2009); heat stabilizers; fillers (Eriksen et al., 2018); anti-microbial additives; and pigments (Dimitrakakis et al., 2009). Limited evidence from Eriksen et al. (2018) and Whitt et al. (2012) indicated that some PTEs may be passed along the value chain as a legacy from their previous use (Table S16). In both studies, the concentrations were reported to be "low" for all elements; however, as indicated, the majority were unlikely to be intentionally added, suggesting that they had originated from a source that was not commensurate with their intended secondary use.

A "low" concentration only provides a partial indication of hazard potential and does not indicate the probability of transfer from the polymer matrix into receptors. Migration and abrasion tests would be needed to confirm this probability, but they were not carried out by Whitt et al. (2012) or Eriksen et al. (2018), meaning that the potential hazard exposure from the concentrations identified was not determined. Nonetheless, Eriksen et al. (2018) pointed out that as the amount of recycling increases, there may be potential for some elements to persist in the value chain and, combined with the addition of metal containing additives, reach levels that could result in undesirable exposure if used in applications such as food contact packaging or toys.

3.4. Other volatile organic compounds

The term "volatile organic compounds" (VOCs) is a coverall for a wide range of substances that evaporate at room temperature and which can be divided into three broad sub-groups (Table S17). Some VOCs are carcinogenic and many have been found to irritate lungs, exacerbate allergies and damage the central nervous system (United States Environmental Protection Agency, 2017; Kwon et al., 2018). VOCs occur throughout the natural environment (Nawrocki et al., 2002) and volatility is not necessarily an indicator of potential hazardousness.

In plastics, the volatility of a substance increases the likelihood of its migration to the surface and subsequent release into the atmosphere or into substrates such as food or liquids via food contact packaging (Even et al., 2019). VOCs may already exist in plastics as intentionally added substances or arise as contaminants picked up during the use phase or through waste management practices (Geueke, 2018). Several factors such as contact with food or oxygen, irradiation, or heat can result in the transformation of additives or of the polymer itself during the production, manufacturing and use phases (Geueke, 2013). The result, is the formation of "breakdown products", inherently lower molecular weight substances that are more mobile and which have a higher probability of migration through the plastic (Bradley and Coulier, 2007).

Tsai et al. (2009) observed three examples of VOC concentrations in air at recycled plastics extrusion facilities in Taiwan which indicate cross contamination of plastics (Table S18). For instance, 1–3 butadiene was detected in air at a PVC waste reprocessing plant at concentrations 7–17 times greater than indoor air reference levels (IARL) proposed by Health Canada (2018). As 1–3 butadiene, is not breakdown product of PVC, the likely cause was contamination of the PVC with ABS. As a result of this cross-contamination, the risk manager of the plant would not be able to anticipate and therefore calculate the exposure to the workforce of this known carcinogen (Sielken and Valdez-Flores, 2015) without constant, costly air emissions monitoring. Without further research, it is not possible to say whether cross-contamination of plastics is common. However, given the scale of the industry, and the potential exposure to carcinogenic substances, it seems a plausible reality that should undergo further investigation.

4. Results: safety challenge 2 - extrusion of secondary plastics

Once plastics have been collected for recycling, they are sorted into polymers and graded before being passed to so-called "reprocessors". There, they are usually (not always) comminuted, before being melted under pressure in an extruder which forces the molten material through a die for direct product production (for example injection molding) or palletization. The heat, between 200 and 300 °C, causes unbound substances within the polymer matrix to become excited and migrate to the surface, from where they may be released into the atmosphere as droplets or gasses if the heat is sufficient (Hahladakis et al., 2018). Heat may also transform substances within the plastic as well as the polymers themselves into "breakdown products" (Geueke, 2018). If unabated, it is inevitable that workers and residents in the vicinity will be exposed to these particles, vapors and gasses, which will be deposited into surrounding environmental media such as soils, dust and sediments (Figs. 3 and S6).

In HICs, negative pressure vacuum systems (local exhaust ventilation) are integrated into extruders which carry away harmful emissions, filtering or treating them as necessary before diluting the remnants in the atmosphere at height (Unwin et al., 2012; Health and Safety Executive, 2013). However, as discussed in the following sections, there is evidence that extraction systems are less commonly used in China due to the additional capital and operational costs involved, resulting in potentially harmful

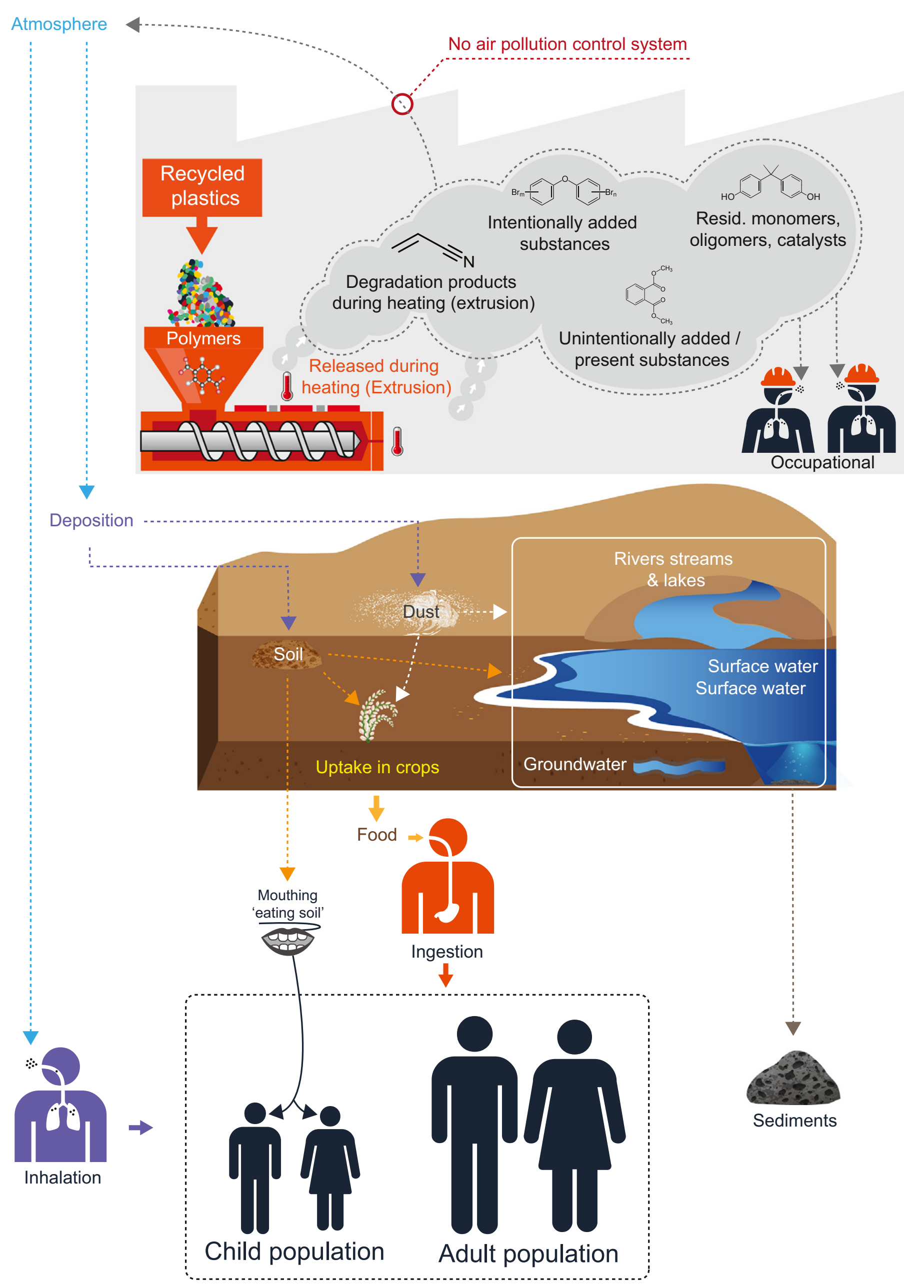

Fig. 3. Hazard exposure conceptual model (hazard–pathway–receptor) associated with extrusion of plastic waste.

exposure of hazardous substances to workers in the sector. Speculatively, the same may be true for other LIMICs, though there is limited evidence to support this.

4.1. Brominated flame retardants

Re-extrusion of plastics that contain BFRs risks exposure to workers; nearby residents; and release of these potentially harmful substances into the surrounding environment where they may persist for many years. Tang et al. (2014) and Tang et al. (2015) determined concentrations of BFRs in soils, sediments and road-dust in a district of China where substantial levels of plastics reprocessing have taken place over recent decades (Table S19). Most samples showed much higher concentrations compared to samples collected from other parts of China and Asia where no plastics reprocessing takes place (Table S20). Neither study was able to categorically determine the source of the BFR contamination; however, the levels identified were consistent with those found in other locations in China where plastics and/or e-waste recycling activities take place. Tang et al. (2014) contextualized their soil and sediment values with analysis of hair samples in the local population, finding higher concentrations in hair of young adults who may be more likely to participate in plastic reprocessing activities. While BFR concentrations in soils, sediments and road dust may result from long-term accumulation in the environment, hair samples are a useful indicator because they indicate existing, ongoing occupational or environmental exposure (that is within the time taken to grow the hair).

The BFR's observed by Tang et al. (2014) and Tang et al. (2015) in soils, sediments and dust may have been transported there via the atmosphere following volatilization during re-extrusion of waste plastics. These could have been sourced either from e-waste or ELV parts that were several decades old, or given that many BFRs are still on the market in South East Asia, relatively new materials. However, given the high boiling points of this class of compounds (~250–450 °C), it seems likely that open burning of unwanted plastic residues may also be an important, and possibly greater source.

BFR degradation mechanisms in the environment are only partly understood and the subject of continuing research (Lassen et al., 2014). The European Chemicals Bureau (2008) suggest a half-life in soils of six months for Tetrabromobisphenol A (TBBPA) which has been shown to break down into mainly bisphenol A, but also tetrabromobisphenol-A-bis(methyl ether), which has a greater potential to bioaccumulate in comparison to TBBPA itself.

Photolysis is thought to be an important mechanism for debromination of PBDEs, particularly the more brominated homologues (Schenker et al., 2008) and Deca-BDE has been reported to have a typical half-life of more than one year in soils. However some studies have shown no degradation at all in sediments under anaerobic conditions after 30 to 40 days (Lassen et al., 2014). Importantly, deca-BDEs undergo debromination into less brominated PBDEs, which have greater potential to accumulate and may have greater toxicity.

While the evidence presented here only covers two studies in one area of China, reprocessing of plastics from e-waste and ELVs is common throughout LIMICs. As regulation and enforcement may be less well-resourced in these countries, it is possible that BFRs represent a substantial risk to extrusion workers and residents living in the vicinity of poorly managed plastics reprocessing facilities worldwide.

4.2. Phthalates

Concentrations of phthalates in air observed by Huang et al. (2013) at ABS and K-resin reprocessors in China were far below the mean long term workplace exposure limits (WEL) recommended by the UK Health and Safety Executive (2020a) (Table 2). Whereas phthalates may expected in ABS (Consumer Product Safety Commission, 2015) and K-resin (Consumer Product Safety Commission, 2016), Yamashita et al. (2009) observed phthalate emissions from extrusion of secondary and virgin plastics where were unlikely to be intentionally added. In each case, the samples analysed by Yamashita et al. (2009) were identified alongside chlorinated compounds suggesting contamination with PVC, which is commonly plasticized with phthalates.

In the study by Wang et al. (2011), water and soil samples from a plastics reprocessing region in China were analysed for phthalate concentration and compared with reference samples. The plastics reprocessing area samples had concentrations orders of magnitude greater than the reference (control) areas indicating that the plastics reprocessing operations were a significant source. To add context, Wang et al. (2011) also analysed blood of occupationally exposed reprocessing workers, concluding that working in the plastics reprocessing industry is a significant independent predictor of higher urinary 8-OHdG (OR = 2.323, $p < 0.01$) for male workers, but not female workers (Table S21).

One other study by Petrovičová et al. (2016) detailed in Table S21, analysed the urine of Slovakian workers in the plastics extrusion sector, finding significantly ($p < 0.02$) higher concentrations of phthalates in those workers compared to waste collectors and the student control group. However, his study did not indicate the type of plastics being extruded or whether they were from recycled feedstock. Therefore there is little relevant conclusive evidence which adds to the present review and the risk to the workers was not modeled. The emissions observed by Huang et al. (2013) and Yamashita et al. (2009) are also concerning because they all indicate cross-contamination of feedstock; however, the concentrations were so low that it is hard to conclude that these are a considerable cause of occupational or public exposure.

4.3. Other volatile organic compounds

VOCs are produced during plastics extrusion due to interactions between the polymer and the various additives, polymerization residues, and unintentionally present substances. They readily evaporate at room temperature, so when plastic is extruded at between 150 °C and 300 °C, any VOCs present begin to be released into the surrounding atmosphere (Hahladakis et al., 2018).

This review identified three studies at 11 plastics reprocessing facilities in China reprocessing nine polymers, each of which had limited or no emission controls; relying instead on dispersion and dilution through open windows and doors to reduce exposure to the workers. The studies analysed atmospheric concentrations of 20 to 30 different VOCs – though only total VOCs are compared in Table S22.

Levels of VOCs in one of the ABS plants and one of the PS plants studied by Mitchell (2015) were very high in comparison to all other facilities investigated. In both cases, styrene dominated the emission profile (data not shown here), representing 63 % (ABS: 630,000 $\mu g.m^{-3}$) and 65 % (PS: 310,000 $\mu g.m^{-3}$) of the total VOCs emitted.

Mitchell (2015) extrapolated their field sampling to model long-term risk from VOC exposure, finding no risk to extrusion workers in the PP, PE and PC plants, but chronic and acute risks in the ABS and PS plants (Table S23). They also sampled air in so-called ‘residential microenvironments’, defined by the authors as being homes in the same building or the same room as the extrusion activities. As with the workers, the hazard index for residents living in close proximity to the PP, PE and PC plants was below one, however emissions of VOCs from PS, PA, ABS and PVC plants would lead to a risk of cancer over their lifetimes.

Laboratory simulations of recycled and virgin plastics extrusion carried out by Yamashita et al. (2009) found higher total VOCs (toluene eq.) emitted by recycled plastic pellets compared to virgin material, the latter of which showed non-detectable quantities of almost all individually VOC species (Table S24). Yamashita et al. (2009) were not able to report the proportions of each polymer in the recycled pellets, limiting potential extrapolation of the results to determine regional or global emissions.

Yamashita et al. (2009) also found that when low-density polyethylene (LDPE) was heated to 250 °C compared to 200 °C, total VOC emissions increased by a factor of ten. While this aspect of the study only investigated one polymer at two temperatures, it indicates that VOC emissions can be controlled by cost-effective process control as well as post-process abatement.

Table 2
Total phthalate concentrations observed in atmospheric samples; plastics samples in the laboratory and in water and soils in China.

Ref.	Context	Sampling			Substance		Units	Conc. Mean		Key findings	USMR[#]
Huang et al. (2013)	Guangdong, CHN	Atmospheric field sampling of emissions at plant	Inside	PC-ABS plant	Total Phthalate	Particle phase	ng. m^{-3}	263.4	SD 23.4	• Very low emissions of phthalates detected – below workplace exposure limits (WEL) over eight hours of 5,000,000 ng.m^{-3} recommended by the UK Health and Safety Executive (2020a) • Exhaust gasses not controlled at plastics extrusion facilities in region • Steps should be taken to provide workers with respiratory protective equipment	K
			Inside	PC-ABS plant		Gas phase		938.8	SD 56.1		
			Inside	Styrene-butadiene copolymer (K-resin) plant		Particle phase		279.7	SD 26.6		
			Inside	Styrene-butadiene copolymer (K-resin) plant		Gas phase		624.7	SD 47.5		
			Outside	PC-ABS plant		Particle phase		94.7	SD 12.0		
			Outside	PC-ABS plant		Gas phase		266.2	SD 25.2		
			Outside	Styrene-butadiene copolymer (K-resin) plant		Particle phase		68.9	SD 11.7		
			Outside	Styrene-butadiene copolymer (K-resin) plant		Gas phase		144.6	SD 14.9		
			Outside	Reference courtyard (20 km distant)		Particle phase		40.9	SD 7.3		
			Outside	Reference courtyard (20 km distant)		Gas phase		27.6	SD 4.1		
Yamashita et al. (2009)	JPN	Atmospheric emissions sampling in laboratory	Virgin [c] LDPE, PP, PS				µg. g^{-1}[d]	ND		• Phthalates observed in very small quantities alongside chlorinated compounds; likely a result of PVC contamination	D
			Mix PE, PP, PS recycled pellet	Heated in air	dimethyl phthalate			2.94	±0.67		
			Mix PE, PP, PS recycled pellet	Heated in air	diethyl phthalate			2.47	±0.65		
			Mix PE, PP, PS recycled pellet	Heated in N_2	dimethyl phthalate			3.14	±1.0		
			Mix PE, PP, PS recycled pellet	Heated in N_2	diethyl phthalate			1.90	±0.30		
Wang et al. (2011)[b]	Hunan, CHN	Field sampling of environmental media	Well water	Exposed	DEHP		µg. L^{-1}	14.2	(0.32–87.70)		L
			Well water	Reference				0.79	(0.28–2.27)		
			Pond water	Exposed				135.68	(0.31–429.89)		
			Pond water	Reference				0.37	(0.23–0.47)		
			Industry wastewater	Exposed				42.43[a]	(0.36–161.86)		
			Industry wastewater	Reference				n/a			
			Agricultural soil	Exposed			µg. g^{-1}	13.07	(0.85–37.23)		
			Agricultural soil	Reference				0.81	(ND–5.81)		

[a] Comparison between exposed and reference concentrations significant ($p < 0.05$).
[b] Samples collected in October 2008.
[c] Unknown additive content.
[d] Expressed as per g of plastic heated.
[#] Uncertainty, strength of knowledge and methodological robustness (USMR) assessed qualitatively. It is assumed that there are no significant concerns unless marked as: D = use of mixed polymer waste samples with unknown proportions means VOC conc. Cannot be attributed to a single polymer; K = Two highly specific waste plastics extrusion plants were studied with different operating temperatures which could influence results: PC-ABS: 230–300 °C; SBC: 200–230 °C; L = samples collected from historical plastics reprocessing area. Not correlated with particular plastics reprocessing operation. Cause of phthalate release not determined. Could be result of open burning of residues; comminution; agitation; or extrusion. Abbreviations: di-2-ethylhexyl phthalate (DEHP); polycarbonate acrylonitrile butadiene styrene (PC-ABS); concentration (conc.).

According to observations by Tsai et al. (2009), Huang et al. (2013) and Mitchell (2015), adequate control measures to protect workers and nearby residents from exposure to VOCs were seldom implemented in plastics reprocessing facilities in China, at the time of reporting; and it is conceivable that this is also still the case throughout many other LIMICs. Given the acute chronic and carcinogenic risk to workers in ABS and PS plants (Mitchell, 2015), and the carcinogenic risk to workers and residents for the PS, PA, ABS and PVC extrusion plants (Tsai et al., 2009; Mitchell, 2015), there is an urgent need to carry out more research to widen the evidence base for these practices across LIMICs. As a way of benchmarking what is feasible, in a context where legacy contamination is absent in a well-regulated and enforced environment with modern equipment, a UK study of ten virgin plastics extrusion plants found very low VOC concentrations in all air samples (Unwin et al., 2012). Though this comparison does not represent like-for-like similitude, the low concentrations in UK plants infers the efficacy of control measures such as local exhaust ventilation and forced mechanical ventilation and dilution.

4.4. Polycyclic aromatic hydrocarbons

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds that are comprised of at least two aromatic rings joined together. They are generally carcinogenic, with a toxic potency indication of 1 ng.m^{-3} BaP_{eq} (benzo(*a*)pyrene equivalent) concentration, leading to 8.7 cases of cancer per million people exposed (Shivani et al., 2019). While atmospheric PAH concentrations are often associated with combustion activities, they may also be emitted as a result of plastics extrusion, as shown by Huang et al. (2013) who analysed air samples at an ABS and K-resin plant in China (Table S25). Inside both plants, levels of PAH with a toxic equivalency >1 ng.m^{-3} BaP_{eq} were observed in both the gas and solid (particle) phase, indicating a potential cancer risk to the workers. Emissions mitigation controls were not in place at either facility, and according to the researchers, it is uncommon to see these anywhere in the region. Equally concerning is that the workers lacked respiratory protection equipment; the last line of defense in the hierarchy of risk control (Hughes and Ferrett, 2016). The detection of high concentrations of PAHs outside the plants studied by Huang et al. (2013) indicate that exposure is not limited to the workforce. In particular, PAHs with a toxic equivalency of 1 ng.m^{-3} BaP_{eq} in the particle phase was observed on the perimeter of the K-resin plant. It was not clear how close the plants were situated to residential dwellings, however in a theoretical example where residents live and or work in close proximity, there is potentially a carcinogenic risk of 8.7 cases per million people (Shivani et al., 2019).

As a comparison to the findings reported by Huang et al. (2013), Unwin et al. (2012) found very low concentrations of PAHs in the air at UK virgin plastic extrusion plants, with the highest being just 0.4 % of workplace exposure limits set by the Health and Safety Executive (Table S25). The data reported by Unwin, though not directly comparable, indicate that extrusion need not be seen as an inherently hazardous activity provided suitable air pollution control measures are in place.

5. Discussion: risk characterization

5.1. Risk characterization for legacy materials in secondary plastics

Despite very limited evidence, we have found research to indicate that BFRs, phthalates and PTEs have passed through recycling systems and into new plastic products manufactured from secondary material. Authors such as Zeng et al. (2022) and Gerassimidou et al. (2022) have highlighted the occurrence of potentially hazardous substances in waste plastics as a result of increased circular economy activity. However, our risk assessment suggests that the risk of such transfer is relatively low for phthalates and PTEs, with only small quantities measured in the few studies conducted (Table 3).

Perhaps more concerning is the presence of BFRs, which were identified in some secondary products at concentrations exceeding POP Directive limits (European Union, 2019). As with some of the studies on phthalates, the data quality for concentrations of BFRs was impeded by lack of detail on sample characteristics and the intended use of materials sampled. Though complete destruction of BFRs in waste plastics is recommended by UNEP and Basel Convention (2020), small concentrations in secondary products may result in negligible exposure depending on the intended use of the products into which they are incorporated. Nonetheless, the presence of each of these three substance groups (phthalates, PTEs and BFRs) in secondary plastics could infer that plastics recyclers in LIMICs and HICs are not adequately controlling supply chains. The paucity of studies implying this supply chain mismanagement should not be interpreted as a mandate for complacency, given the potentially harmful nature of these substances. Rather, the lack of information on this topic indicates a clear need for further investigation into the presence of these substances in materials containing secondary plastic content.

The greatest risk identified from legacy substances resulted from a lack of stringency by a reprocessor who allowed materials such as PVC and ABS to be co-processed with other plastics that would not otherwise emit hazardous substances when extruded (Tsai et al., 2009). As a consequence, several carcinogenic VOCs were detected there at high concentrations. Despite that this risk was highlighted only by a single study in Taiwan, the lack of emissions controls reported to be implemented in many LIMICs indicates vulnerability to a potentially large workforce and just states the insufficient attention and research on the topic.

5.2. Risk characterization for extrusion of secondary plastics

We considered it likely that the majority of reprocessors in HICs have measures in place to control emissions and protect their workers from exposure to potentially hazardous substances. Under those conditions, in the wider global context, the risk to human health is assumed comparatively trivial and therefore we did not assess the HIC context.

In some LIMICs, we found indications that the implementation of such control measures is less consistently applied, with some facilities relying on passive ventilation to dilute emissions and a lack of provision of respiratory protective equipment to the workers (Table 3). Clearly the extrusion of some polymers will result in an emission profile that is more hazardous in comparison to others, but consistently, PS, PA, ABS and PVC all featured as having a greater likelihood of producing hazardous emissions. It happens that PC-ABS and K-resin were identified during the search in the present study, however although the amount of material being processed (plant throughput) could not be verified in this study, it is certainly less than the polyolefins, PET, PS and PVC.

Though we assess the non-occupational risk of VOC exposure to be slightly lower than the occupational, it is worrying that our risk assessment indicates that plastics reprocessing workers in some LIMICs may have a similar risk of exposure to hazardous emissions as residents who live nearby. This was only supported by a single study (Mitchell, 2015), which reported that the long-term exposure from ABS and PVC was enough to rate the non-carcinogenic risk to those residents as being “very high”. However, the level of proximity was described loosely by Mitchell (2015) who used the term “near” without specifying the distance, only indicating that sampling was carried out in “residential microenvironments”; other rooms within the same building or in the workshops themselves. The level of ventilation and number of rooms separating residents from the extrusion activities was not stated. Without this context, the level of attenuation, dispersion and dilution was unclear.

It is not known how many people live near, or in the same building as secondary or primary plastics reprocessors, and therefore the size of the globally exposed population. However, given that these emissions are relatively straightforward to mitigate, and that residents often have no choice but to sustain exposure, there appears a compelling case for further air quality research in proximity to plastics reprocessors. This would determine

Table 3
Risk characterization summary for legacy substances and extrusion of secondary plastics.

Haz.	Pathway	Receptor	Geog.	Evidence and justification for risk assessment	Notable material/ polymer/ substance	Uncertainty (aleatoric and epistemic)	Receptor vulnerability	L	S	R	Global receptor context
Hazards from legacy substances in secondary plastics											
BFR	Mouthing, abrasion, breathing and dermal contact	Children Population	ITA, CZE, DEU, CHN, TUR, NLD, DNK	• Multiple examples of BFRs re-entering product streams through secondary plastics, however most concentrations were very low (Chen et al., 2009; Lyu et al., 2015; Leslie et al., 2016; Guzzonato et al., 2017; Pivnenko et al., 2017). • Five samples of toys and two samples of food contact items had levels of BFRs that exceed revised POP Directive (European Union, 2019) threshold of 500 μg.g^{-1} (Guzzonato et al., 2017). • Evidence indicates that at levels identified (Chen et al., 2009; Lyu et al., 2015), exposure and non-carcinogenic risk is likely to be very low; however future research should establish if higher concentrations (Hong-Gang et al., 2016; Pivnenko et al., 2017) are prevalent.	• Polymers: PS (Hong-Gang et al., 2016; Pivnenko et al., 2017), ABS (Pivnenko et al., 2017) • Products: ELV parts (Leslie et al., 2016), WEEE (Leslie et al., 2016), Toys (Guzzonato et al., 2017)	• Robust analysis of concentrations but uncertainty over: a) The prevalence of BFR contamination in secondary products b) The intended use of secondary pellets.	• Children are more vulnerable to exposure due to lower body weight and propensity for mouthing	2	4	8	All plastic product consumers LIMIC / HIC
Phth.	Mouthing, abrasion, breathing and dermal contact	Children Population	CHL, USA CHN, DNK, DEU, NLD	• Although modelling (Lee et al., 2014) indicates that phthalate exposure to two year olds in Denmark may be increased by the use of secondary materials, there are only a limited number of studies that indicate high levels re-entering the product stream in secondary plastics in amounts which result in a high concentration. • High concentrations not found in high risk products such as those used in food contact items or toys.	• Non-specified plastics from source separated collection (Pivnenko et al., 2016) "roadside trash" in Chile (Simoneit et al., 2005)	• Very few of the samples analysed reported the specific polymer or product type and in many cases products and polymers were mixed, making it difficult to determine the polymer or product which included the phthalate content.	• Children are more vulnerable to exposure due to lower body weight and propensity for mouthing	1	4	4	All plastic product consumers LIMIC / HIC
PTE	Mouthing, abrasion, breathing and dermal contact	Children Population	USA CHN, DNK	• Though Whitt et al. indicated that the Mn arose from contamination, two studies (Whitt et al., 2012; Eriksen et al., 2018) concluded that PTEs were unlikely to be a consequence of contamination of secondary materials. • Metal content was higher in waste samples compared to virgin samples in one study (Eriksen et al., 2018) but still at levels far below low RoHS Directive and California (Toxics in Packaging Prevention Act, 2005) thresholds.	• None	• Migration/abrasion testing was not performed which means that exposure through food contact or mouthing was not assessed.	• Children are more vulnerable to exposure due to lower body weight and propensity for mouthing	1	4	4	All plastic product consumers LIMIC / HIC
VOC	Atmosphere/ inhalation	Plastics reprocessing workers	CHN	• Volatile organic compound emissions identified in air at plastics extrusion plants in Taiwan (Tsai et al., 2009) indicating that: - PVC had been contaminated with ABS waste - PP and PE had been contaminated with PVC waste. • Levels of 1-3 butadiene (carcinogen) 7-17 times more than the IARLs set by Health Canada, posing a risk to workforce.	• PVC / ABS (Tsai et al., 2009)	• n/a	• Provision of air pollution control measures rare in LIMICs • Respiratory protective equipment (RPE) may not be provided	2	5	10	Recycled plastics extrusion workers LIMIC
Hazards from extrusion of secondary plastics											
BFR	Atmosphere/ inhalation	Plastics reprocessing workers	CHN	• Evidence for BFR emissions from extrusion is through large concentrations (Tang et al., 2014) observed in soil and sediment which may have been deposited there from the atmosphere; and originated from extrusion activities. • Concentrations in dust (Tang et al., 2015) may have arisen from extrusion, burning or abrasion of plastics debris on the road. • Concentration in hair samples higher in young people indicating occupational exposure (Tang et al., 2014).	• Extrusion of plastics recovered from WEEE, ELVs where BFRs historically used as additives.	• Atmospheric samples not obtained so evidence is limited to concentrations observed in soil, sediment and hair.	• Provision of air pollution control measures rare in LIMICs. • RPE may not be commonly/ consistently provided in LIMICs.	3	4	12	Recycled plastics extrusion workers in LIMICs
	Atmosphere/ inhalation; soil/uptake in food	Population living nearby to extrusion					• Provision of air pollution control measures rare in LIMICs. • Emissions controlled through dispersion and dilution in ambient atmosphere.	3	4	12	Residents living near extrusion plants in LIMICs
Phth.	Atmosphere/ inhalation	Plastics reprocessing workers	CHN, JPN, SVK	• Workers in PC-ABS and K-Resin plants exposed to levels of phthalates that are 4-25 times greater than the reference (Huang et al., 2013). • Blood (Wang et al., 2011) and urine (Petrovičová et al., 2016) samples from plastics reprocessing workers in China and plastics workers in Slovakia respectively indicate significantly higher exposure to phthalates.	• Evidence from PC-ABS and K-Resin plants, however logically PVC extrusion plants are also at very high risk.	• Aside from the PC-ABS and K-Resin plants, the types of plastics which workers have been exposed to are not recorded.	• Provision of air pollution control measures rare in LIMICs. • RPE may not be commonly/ consistently provided in LIMICs.	3	4	12	Recycled plastics extrusion workers in LIMICs
VOC	Atmosphere/ inhalation	Plastics reprocessing workers	CHN	• Total VOCs in PS and ABS plants (Mitchell, 2015) were very high in comparison to the other plants, being mostly comprised of styrene and resulting in acute chronic risk to their workers. • VOCs in the PS, PA, ABS and PVC plants also result in carcinogenic risk to workers.	• Very high risk: PS and ABS. • High risk: PS, PA, ABS and PVC.	• Further analysis needed to assess the risks from individual polymers.	• Provision of air pollution control measures rare in LIMICs. • RPE may not be commonly /consistently provided in LIMICs.	3	5	15	Recycled plastics extrusion workers in LIMICs
	Atmosphere/ inhalation; soil/uptake in food	Population living nearby to extrusion		• Carcinogenic risk to residents for the PS, PA, ABS and PVC (Mitchell, 2015).	• High risk: PS, PA, ABS and PVC.		• Provision of air pollution control measures rare in LIMICs. • Emissions controlled through dispersion and dilution in ambient atmosphere.	3	4	12	Residents living near extrusion plants in LIMICs
PAH	Atmosphere/ inhalation	Plastics reprocessing workers	CHN	• Levels of PAH in the PC-ABS and K-Resin plants have a toxic equivalency greater than 1 ng.m^{-3} BaPeq in both the gas and particle phase, indicating a significant cancer risk to the workers (Huang et al., 2013).	• High risk: PC-ABS and K-Resin.	• Aside from the PC-ABS and K-Resin plants, the types of plastics which workers have been exposed to are not recorded.	• Provision of air pollution control measures rare in LIMICs. • RPE may not be commonly/ consistently provided in LIMICs.	3	5	15	Recycled plastics extrusion workers in LIMICs

Abbreviations: likelihood (L); severity (S); risk (R); hazard being assessed (Haz.); phthalates (Phth.); geographical context (Geog.); potentially toxic elements (PTE); polyethylene (PE); polypropylene (PP); polyvinyl chloride (PVC) low income and middle income countries (LIMIC); high income countries (HIC); acrylonitrile butadiene styrene (ABS); volatile organic compounds (VOC); phthalates (phth.); brominated flame retardants (BFR); indoor air reference levels (IARL); end of life vehicle (ELV); waste electrical and electronic equipment (WEEE); respiratory protective equipment (RPE); polycarbonate/acrylonitrile-butadiene-styrene (PC-ABS); styrene-butadiene copolymer (K-Resin); polystyrene (PS); polyamide (PA); benzo(*a*)pyrene equivalent (BaPeq).

whether the public health risk highlighted by Mitchell (2015) is an outlier, or whether it indicates a threat to a much larger population.

As with many other hazards, the concentrations in soils, sediments, dusts and in the hair of exposed subjects provided circumstantial evidence (Tang et al., 2014; Tang et al., 2015) of exposure to potentially hazardous substances from extrusion; being collected in historically active reprocessing areas in China. However, whether these arose from open burning (Velis and Cook, 2021), extrusion and/or abrasion can only be speculative with the low level of available evidence.

6. Conclusions and outlook

The attention placed on circular economy principles and practice in recent decades has resulted in a drift of focus from the formative driver for modern waste management, which was to protect public health and safety. Here, we report for the first time (answering RQ1), a global systematic review of evidence that indicates harm to human health for those who work with waste plastics and those who are affected by plastic waste processing activities, including the controlled operations of plastics reprocessors. We derived prevalent risk scenarios of hazard-pathway-receptor combinations (answering RQ2). These were mapped into a conceptual flow and then ranked according to the indicative risk to human health, allowing us to indicate priorities for future research agenda.

The presence of legacy VOCs, BFRs, phthalates, and PTEs in some plastic products sold in HICs indicates that the recycling part of our circular economy does not necessarily provide for safe and final sinks for substances of concern in plastics (Kral et al., 2013; Johansson et al., 2020; Stanisavljevic and Brunner, 2021). In most cases, concentrations were far below limits imposed by the EU regulations / standards (for instance). But their presence indicates that even with the most stringent risk management systems available, for instance in Europe, that substances from the previous use have the potential to persist in new plastic products, from where they could migrate into the environment (and disperse) and potentially harm human health.

Heating recycled plastics, for instance during extrusion, exacerbates the release of both intentionally and non-intentionally added substances into the environment, and increases the risk of them entering the human body. As the global circular economy for waste plastic proliferates as we anticipate, the risk of non–traceable hazardous materials and substances re-entering the use-phase is likely to increase. This could result in increased dispersion of substances of concern all over the world. Our evidence indicates that plastics reprocessing workers in some parts of the Global South are not protected from occupational exposure to atmospheric pollutants. Such risks are exacerbated if the provenance of the plastics to be recycled cannot be reliably determined, which is often the case, particularly in the Global South.

Given the paucity of current information on the topics reviewed here, and the potential for harm to human health and increase mortality, it is recommended that further studies are carried out in small-scale and rudimentary plastics reprocessing facilities to determine: 1) The potential and actual exposure to potentially hazardous substances sustained by plastics extrusion workers; 2) The content of non-intentionally and intentionally added substances of concern in feedstocks across the plastics recycling value chain linked to reprocessing feedstock provenance; and 3) The efficacy of different approaches to managing emissions from extrusion, especially rudimentary, to assist with developing a road map towards safer working practices in resource-scarce contexts. It is hoped that our research will serve as a baseline evidence summary for the aforementioned future studies.

CRediT authorship contribution statement

Ed Cook: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Resources, Validation, Visualization, Writing – original draft, Writing – review & editing. **Michiel Derks:** Formal analysis, Data curation, Investigation. **Costas A. Velis:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing.

Data availability

All data associated with this review can be found within the main manuscript and the Supplementary Material, and original sources are stated

Declaration of competing interest

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Appendix A. Supplementary data

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