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

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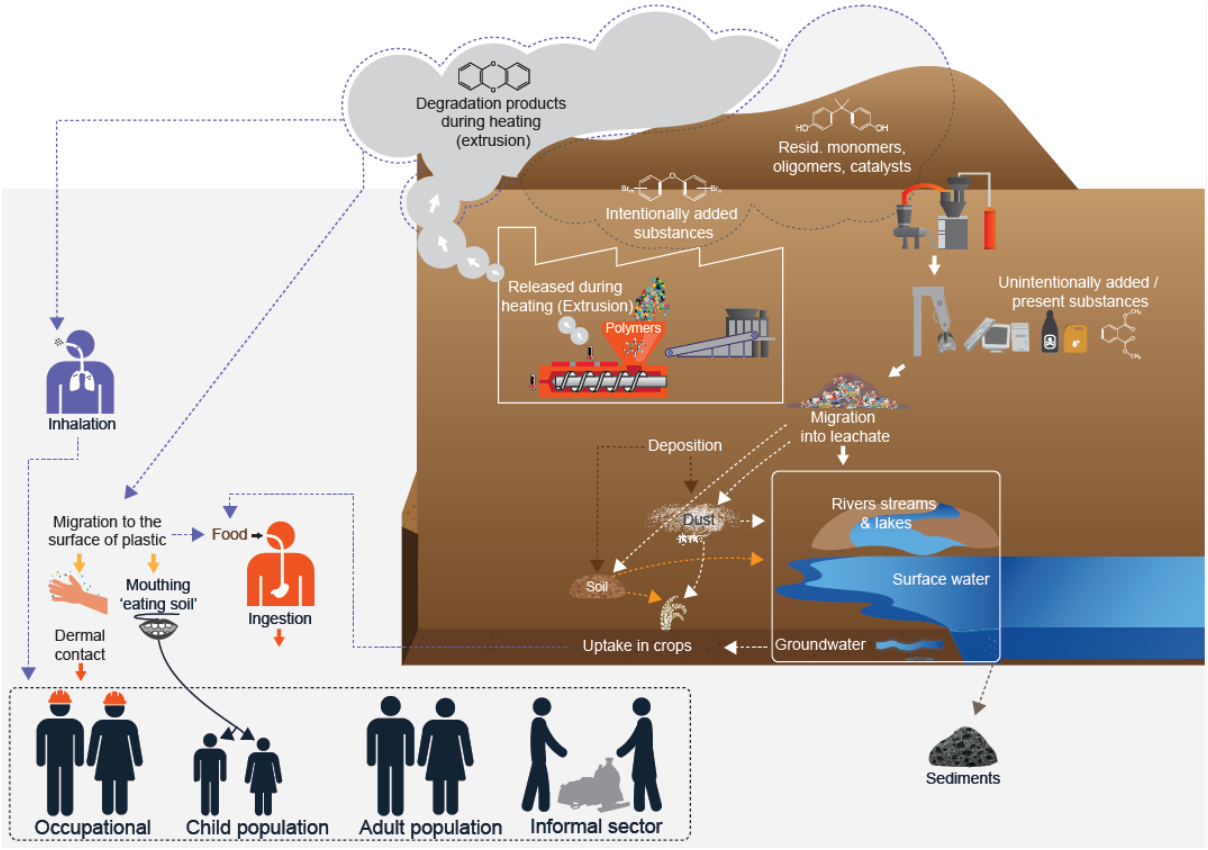
Abstract

Increasing aspirations to develop a circular economy for waste plastics will result in an expansion of the global plastics reprocessing sector over the coming decades. Here we focus on two critical challenges within the value chain that as a result of such increased circularity may exacerbate existing issues for occupational and public health (1): Legacy contamination in secondary plastics, addressing the risk of materials and substances being inherited from the previous use and carried through into new products when the material enters its subsequent use phase; and challenge (2): Extrusion of secondary plastics in reprocessing, an end process of conventional mechanical recycling of plastics, involving heating secondary plastics under pressure until they melt and can be formed into new products. Via a systematic review (PRISMA guidelines, adapted), we considered over 4,000 sources of information, refined and consolidated into 20 relevant sources, which were critically assessed. We also derive prevalent risk scenarios of hazard-pathway-receptor combinations, subsequently being ranked. Our critical analysis highlights that despite stringent regulation, industrial diligence and enforcement, occasionally small amounts of potentially hazardous substances are able to pass through these safeguards and re-enter in the new product cycle. Although many are present at concentrations unlikely to pose a serious and imminent threat, their existence may be an indication of a wider or possibly increasing challenge of pollution dispersion, as the plastics reprocessing sector proliferates. But, in the Global South context, such controls may not be in place. Several studies showed emission control by passive ventilation, through open doors and windows followed by dilution and dispersion in the atmosphere, resulting in increased occupational exposure. It is recommended that further investigations are undertaken to establish the scale and magnitude of such phenomena, especially given the limited evidence base, with results informing improved future risk management protocols of a circular economy for plastics.

Keywords: Plastic, Solid waste, Health and safety, Risk, Global South, Resource recovery, Circular economy

Graphical Abstract



Abbreviations

8-OHdG	8-hydroxy-2′-deoxyguanosie
ABS	acrylonitrile butadiene styrene
BaPeq	benzo(a)pyrene equivalent
BBP	benzyl butyl phthalate
BDEs	brominated diphenyl ethers
BFR	brominated flame retardants
bw	body weight
CAGR	compound annual growth rate
Conc.	concentration
DBP	dibutyl phthalate
DEHP	di(ethylhexyl) phthalate
DEP	diethyl phthalate
DMP	dimethyl phthalate
EfW	energy from waste
ELV	end of life vehicle
EPS	expanded polystyrene
EU	European Union
Geog.	geographical context
Haz.	hazard
HBCD	hexabromocyclododecane
HC	hydrocarbons
HDPE	high density polyethylene
HH	household
HIC	high income countries
HSE	Health and Safety Executive
IARL	indoor air reference levels
K-resin	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
SBC	styrene-butadiene copolymer
SD	standard deviation
SOD	serum superoxide dismutase
SVOC	semi volatile organic compounds
TBBPA	tetrabromobisphenol A
temp.	temperature
TEQ	toxic equivalency
tonne	1,000 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

1. Introduction

Despite the great benefits engineered polymers (plastic materials and products) bring to society, plastics may also have substantial drawbacks, especially when they have served the purpose for which they were originally intended (Burns and Boxall, 2018; Rochman et al., 2016). Management of plastic waste is a pervasive, multifaceted and highly debated challenge of our times (Haward, 2018), and despite the recent calls and laws on limiting production of single use/ disposable plastic articles (da Costa et al., 2020; Xanthos and Walker, 2017), the consumption of plastics and hence plastic waste generation continues to follow an exponential growth curve (Geyer et al., 2017). 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.
- **Second**, a recent imperative for a circular economy for plastics is gradually being established, exploring how resource recovery from waste can be extended to include re-use, remanufacturing, refurbishing, along with waste avoidance and minimisation.
- **Third**, non-negligible quantities, approximately 6 million tonnes in 2018 (United Nations, 2020), of plastic waste (used/ secondary) are traded internationally; a market that has represented an integral component of the global circular economy for several decades (Velis, 2014), characterised by exports of often unsorted mixed plastics from high income countries (HICs) to low 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 recycling these materials are being mismanaged (and may also be leaking into the aquatic environment) (Secretariat of the Basel Convention, 2019).

In addition, the globalised manufacturing of both primary and secondary plastics, could in principle, result in the dispersion of any chemical substances of concern, due to variable levels of quality assurance, legal provisions and enforcement, and overall control in the manufacturing process (Johansson et al., 2020). The recent 'National Sword' policy and operation by Chinese authorities (often also called 'China ban') on plastic scrap imports (Ministry of Ecology and Environment, 2017), and the amendment of the Basel Convention (Secretariat of the Basel Convention, 2019) to classify some types of plastic waste as hazardous denote a rapidly evolving policy landscape, impacting on market realities.

Most importantly, 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 economies, plastic waste is managed by being disposed of in landfill, recovered as fuel in energy from waste (EfW) plants, or mechanically recycled. Yet, this formal waste industry collectively has one of the highest accident rates of all industrial sectors (Doherty, 2019; Health and Safety Executive, 2018) in many countries and although disaggregation of safety reporting data is problematic, plastics represent a significant proportion of the composition of waste (Kaza et al., 2018).

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 estimated at 18 and 49 million tonnes per annum respectively (Lau et al., 2020). Virtually all of the material collected for recycling in LIMICs is carried out by waste pickers (Cook and Velis, 2020); informal entrepreneurs who may number between 10 and 20 million (Lau et al., 2020; Wilson et al., 2015). 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 and Mavropoulos, 2016; Velis et al., 2009); a goal largely achieved in the Global North as a result of investment and technological advances. 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. However, as we have just demonstrated, waste related risks persist in parts of the world, and potentially affect everyone via globalised secondary supply chains, our shared atmosphere, and our delicate ecosystems.

The system through which plastic waste flows across society and the environment (**Figure 1**), is in many ways similar to other major constituents of solid waste, but for the complexity of the many thousands of polymer and additive combinations and their persistent and fragmentary behaviour in the environment when mismanaged. Whereas the system itself has been the subject of serval global and national studies, for instance Geyer et al. (2017), Lau et al. (2020), and Bai et al. (2018), surprisingly or not, there have been few systematic efforts to quantify and compare the related risks between human health and ‘plastic waste’.

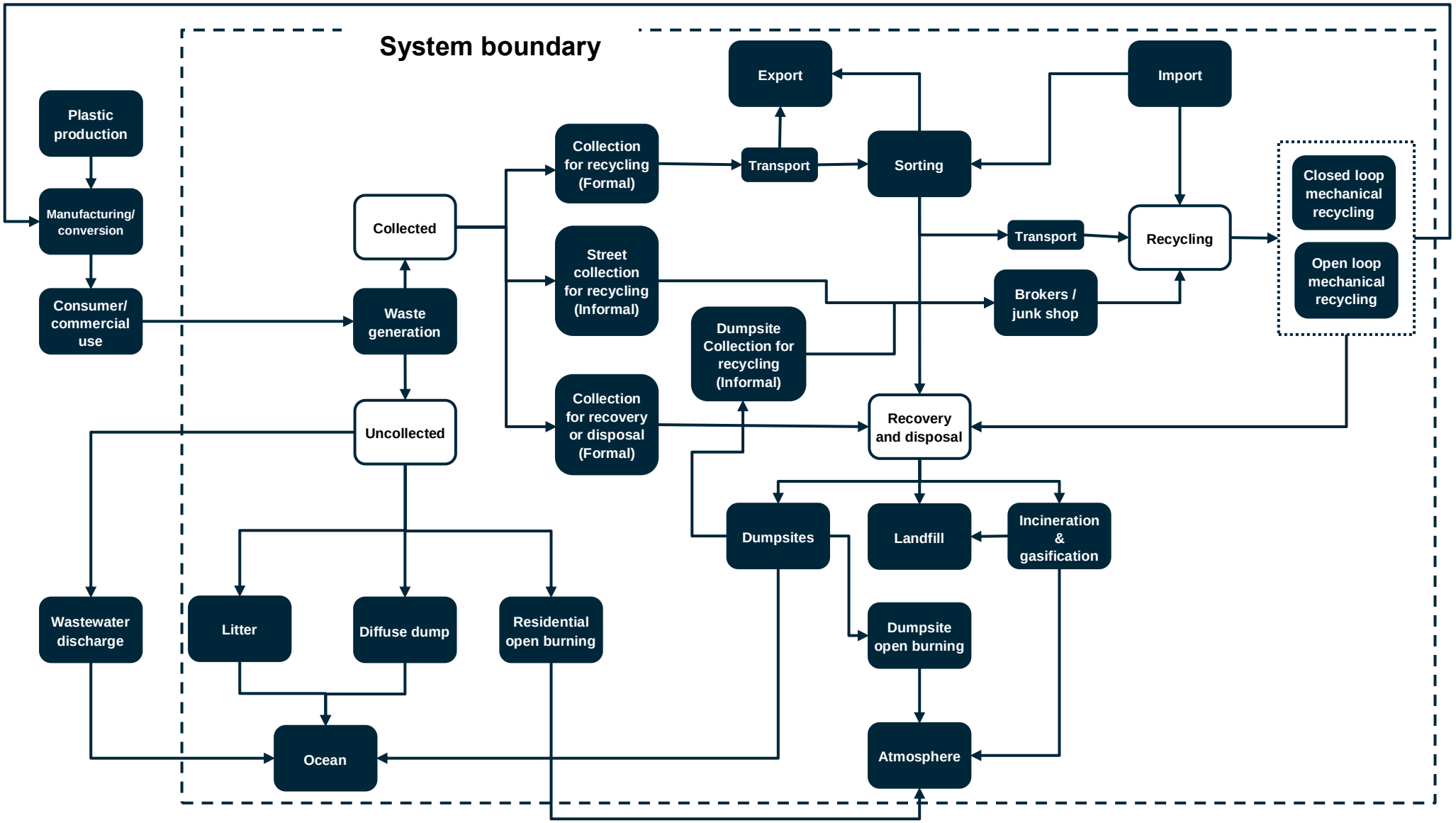


Figure 1: Generalised material flow of plastic and plastic waste management in society.

Several general reviews of solid waste management exist that summarise health and safety challenges (Ferronato et al., 2019; Giusti, 2009; Searl and Crawford, 2012). Although they include 'plastic waste' as a component of the overall 'solid waste' category, it is one of the least focussed materials, being relatively benign in comparison to other hazards such as pathogen infection or road traffic related incidents. Two exceptions (Azoulay et al., 2019; Williams et al., 2019) exist in the grey literature. Williams et al. (2019) focussed their efforts 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 this estimate. 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 plastic waste, identifying pathways but without determination of potential exposure. Hahladakis et al. (2018) provide an extensive review of the additives in plastics materials and their fate, but without an overall risk-based framework and quantification.

Beyond these, there are multitude studies, many of which are reviewed here, that focus on individual substances arising in relation to plastic waste processing or mismanagement such as brominated flame retardants, phthalates, and potentially toxic elements. Though, we have not found a harmonised, systematic approach that has addressed all of the issues related to plastic waste in a single consistent manner. Even in these tightly focused studies on substances of concern, the lack of coherent harmonised approaches on risk and hazard, make the comparison of any research outputs a highly debatable exercise. For example, differentiating hazard from risk and quantifying hazard exposure is challenging, not only for decision makers, but also for scientists and researchers who, despite their best efforts, do not always have the time and resources to reach definitive answers.

Therefore, here we bring some of the above concerns into focus by ways of systematic reviewing, consolidating and analysing the available literature to provide a balanced and pragmatic overview of the most significant human health risks that could be associated with plastic waste processing. We group the information into key thematic areas dealing with (i) legacy substances present in plastic materials; and (ii) prevalent (manual/mechanical) recycling operations. Notably, 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 (2020). 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 as, we suggest, that they belong in another in-depth study.

2. Methods

A brief summary of the methods is provided here, with more comprehensive details presented in **Section S.1** of the **Supplementary Information**.

2.1. Systematic review

A systematic review (**Section S1.2)** explored three research questions (**RQ**) following the principles of the PRISMA guidelines as suggested by Moher et al. (2009):

- **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?
- **RQ3:** What research could be carried out that would have the greatest impact on harm reduction in the plastic waste management sector?

Some adaptations to PRISMA were necessary as the RQs required the review of a wide range of evidence across a complex sector rather than answering a single tightly defined question. As recommended by Gusenbauer (2019), we searched Scopus, Web of Science and Google Scholar to improve the probability of capturing all literature (**Section S.1.1.5**). Boolean search terms were tested with one at a time sensitivity analysis to ensure that they captured the maximum number of relevant papers (**Section S.1.2**). These were screened by a single reviewer according to pre-defined criteria and periodically checked by a second reviewer to ensure a consistent approach (**Section S.1.4**). Further searches were carried out such as snowball and citation searching (Cooper et al., 2018). Online datasets and libraries were also searched, from organisations 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) (HSE) in the UK. The results of this review are summarised in **Section S.1.6.**

Information sources were categorised by the type of waste management activity and these were further distilled into three overarching activity based categories. The first two, 'legacy materials' and 'extrusion' are reported in the present manuscript and the 'open burning' category is reported in a separate manuscript presented by Velis and Cook (2020). The methods employed in each study being reviewed were qualitatively assessed to ensure that a robust scientific approach had been taken and those what appeared flawed were excluded. Moreover the risk of bias was addressed qualitatively in the narrative where appropriate, however this was not addressed according to the structured approach recommended by Moher et al. (2009).

2.2. Uncertainty, strength of knowledge and methodological robustness (USMR)

The strength of information reviewed in each of the sources was assessed as required by PRISMA guidelines(Moher et al., 2009). Where appropriate, information was qualitatively coded on a case-by-case basis according to 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, the pathway through which the hazard may reach a receptor and the receptors themselves. This allowed the creation of source-pathway-receptor diagrams which enabled the visualisation of risk in each of the three activity groups. These are shown in each of the **Sections 3** and **4,** whilst a combined version, although it represents a partial 'result' is shown here in the method to assist understanding in **Figure 2**.

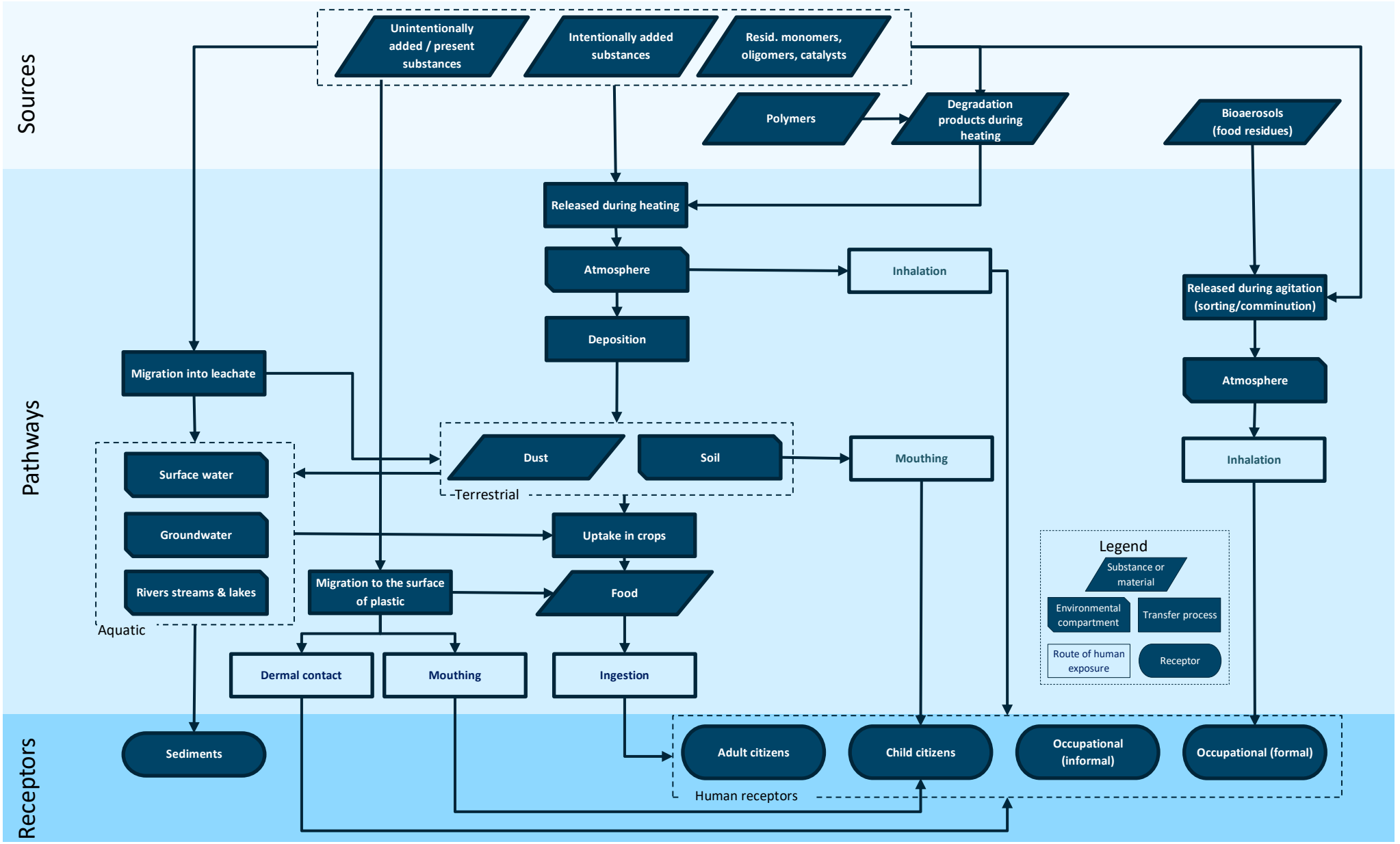


Figure 2: Hazard exposure conceptual model (source–pathway–receptor) for waste plastics.

2.1. Stakeholder engagement

A system flow diagram was used to identify the types of stakeholders in the plastic waste system who may be able to provide primary information or indicate secondary sources of information relevant to the research questions (**Section S.1.7**). Once identified, stakeholders were provided with a preliminary information booklet and questionnaire. Semi-structured interviews (45-60 minutes) were then carried out to obtain further information, the results of which are presented, where relevant as personal communications.

2.2. Risk based approach

As many of the studies measured non-comparable data, a meta-analysis was not appropriate. Therefore as an approach to summarising and ranking risk to human health, a semi-quantitative method (**Section S.1.8**) 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. As suggested by Kaya et al. (2018) it should be noted that this method is not intended to quantify risk associated with the identified hazard-pathway-receptor combinations or inform decisions directly. Rather it is intended to support decision-making and indicate where efforts for intervention or further research might be directed.

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 was assigned a likelihood and severity score by the project team according to criteria detailed in **Table S 3** and **Table S 4** and these were checked by a Technical Advisory Group of sectoral experts. The product of the likelihood and severity resulted in a colour coded risk score, described qualitatively in **Table S 6**.

Table 1: Matrix used to calculate the relative risk of each hazard-pathway-receptor scenario.

			Consequence				
			Very slight	Slight	Moderate	Severe	Very severe
			1	2	3	4	5
Likelihood	Very unlikely	1	1	2	3	4	5
	Unlikely	2	2	4	6	8	10
	Likely	3	3	6	9	12	15
	Very likely	4	4	8	12	16	20
	Inevitable	5	5	10	15	20	25

3. 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. Plastic additives that are transferred through the material chain in this way are referred to as 'legacy additives' (Wagner and Schlummer, 2020), however here we use the term 'legacy substances' (add citation), 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;
- Residual **substances from primary plastic production** such as unreacted monomers, catalysts and oligomers;
- **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 categorised 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); 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).

In many cases these legacy substances are either benign or occur in very low quantities; posing little risk to human health (Wagner and Schlummer, 2020). Even when some

substances occur in larger quantities or are potentially hazardous to health, if they are bound to the polymer or have low migration potential, then they may have limited potential to transfer from the surface and into surrounding media such as food or human skin.

In **Figure 3**, the potential for hazard exposure from legacy substances is illustrated in a conceptual diagram. The arrows represent the pathways through which potentially hazardous substances may move and come into contact with people and the route of exposure.

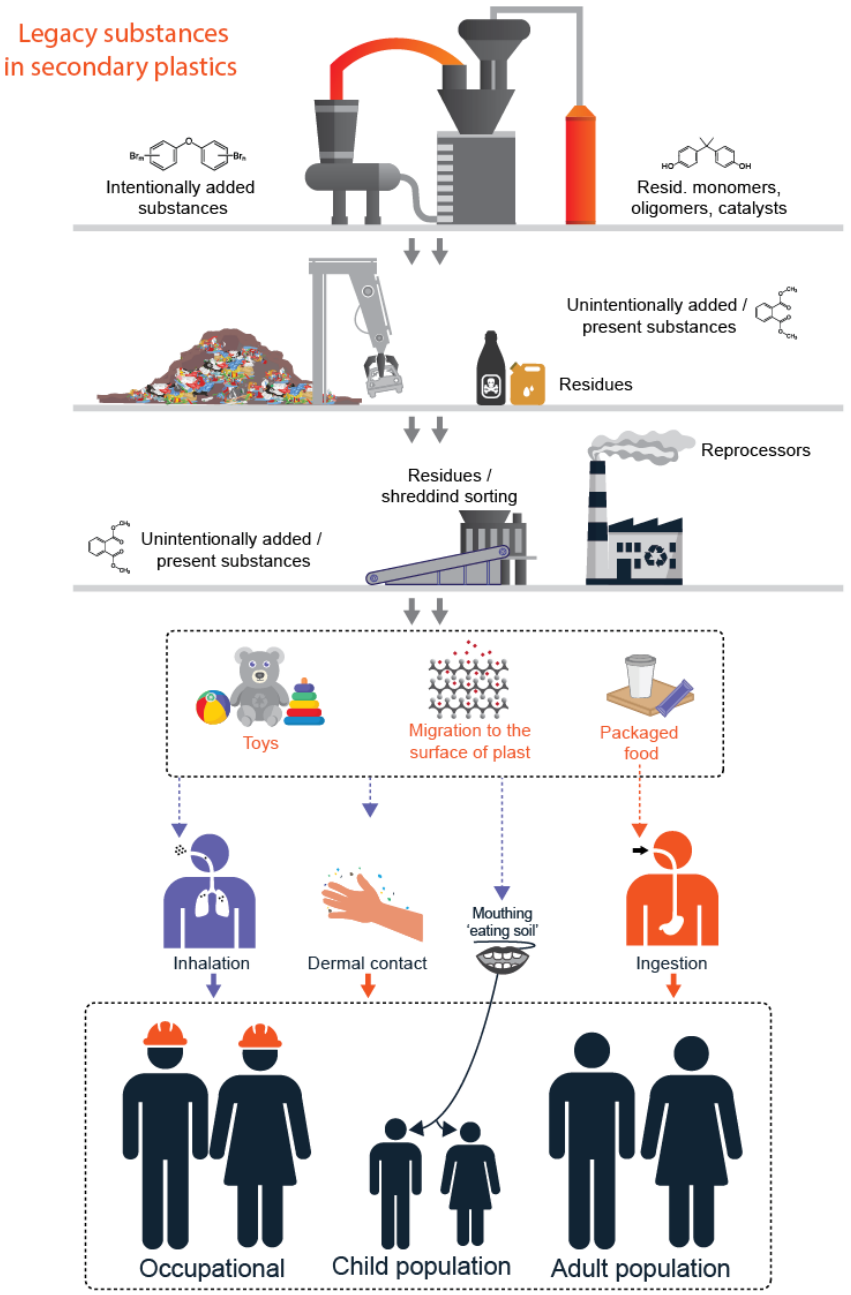


Figure 3: Hazard exposure conceptual model (source–pathway–receptor) associated with legacy substances in waste plastics.

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 modelled risk to human health from these substances.

3.1. Brominated flame retardants (BFR)

This group of substances, bromophenols, hexabromocyclododecane (HBCD), polybrominated diphenyl ethers (PBDEs) (hereafter BFRs) are added to plastics to inhibit combustion chemistry. Many BFRs have been in commercial production since the 1950s, and can be found in automotive, electrical, aeronautical and furnishing applications worldwide where they are expected to continue to arise in considerable quantity for at least the next decade (Covaci et al., 2011; Sharkey et al., 2020; Wagner et al., 2019).

Certain BFRs 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); in particular, the class of brominated flame retardants called polybrominated diphenyl ethers (PBDEs). Historically three major formulas of PBDEs have been in use: Penta-BDE, Octa-BDE and Deca-BDE, between them comprising 209 congeners. Accumulation of specific BFRs in the environment is well documented (Covaci et al., 2011); and the Penta-BDE and Octa-BDE formulations were classified as persistent organic pollutants by the Stockholm Convention in May 2009 (Tang et al., 2014), and the Deca-BDE was added in 2017 (UN Environment, 2017).

The production of both Penta-BDE and Octa-BDE products is banned in the United States (US) (Venkatesan and Halden, 2014) and Europe, however production still continues elsewhere, including in China (Tang et al., 2014); meaning the two congener groups are still at risk of entering the environment. PBDEs are not used in food packaging, as there is little need for flame retardant properties; however, their use in durable goods, such as furniture, clothing, and electrical goods means that many products containing PBDEs remain in use today and are likely to continue to do so for many years to come.

While production continues in China (Yu et al., 2016), BFRs are all but banned in new products (Alaee et al., 2003) in Europe and the US. 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 1,000 µg 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 1,000 µg g^{-1} cannot be recycled until their PBDE and HBCD content has been destroyed or irreversibly transformed. At the time of writing, a 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 contextualise 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 2**). This review identified six research outputs reporting BFRs in varying quantities in new plastic products, plastic wastes, and recycled plastics. Overwhelmingly, the materials and products tested did not contain levels of BFRs that exceeded the RoHS Directive threshold of 1,000 µg g^{-1} plastic. However, the presence in such a wide array of products clearly indicates cross-contamination of secondary plastics streams with legacy BFRs.

Table 2: 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[#]
Chen et al. (2009)	CHN[a]	Assorted toys[b]	ΣPBDE	53		Median conc. in hard toys > others toys all <WEEE and RoHS Directives (1000 µg g^{-1}) Except single sample 5,344 µg g^{-1}	
			ΣDBDPE	5.54			
			ΣBTBPE	0.101			
			ΣPBBs	0.0279			
Guzzonato et al. (2017)	Purch: ITA, CZE, DEU; Man'f: CHN TUR[c]	Rubik's cube		328.1		BFRs found most frequently in toys 1/3 food contact items contained Br 61% contained Br 45% contained ΣdecaBDE >1,000 µg g^{-1} Food contact articles sold on European market are not produced exclusively with food-grade polymers, contravening Regulation (EC) no. 202/2014 Many conc. >1,000 µg g^{-1} contravening WEEE & RoHS Directives	
		Toy gun		4,352.7	*		
		Spring car		1,303.8	*		
		Spring car		944.4	†		
		Car launcher		9,225.8	*		
		Miniature car		284.3			
		Miniature car		1,279.8	*		
		Spring gun		210.5			
		Thermal cup		778.8	†		
		Thermal cup		775.2	†		
		Radio back panel	decaBDE	5,118.8	*		
			ΣdecaBDE	~200-10,000			
		Rubik's cube		386.8			
		Toy gun		661.3	†		
		Spring car		774.4	†		
		Spring car		278.1			
		Car launcher		7747.1	*		
		Miniature car		927.2			
		Miniature car		208.4			
		Spring gun		513.9	†		
		Thermal cup		442.8			
		Thermal cup		471.3			
		Radio back panel	TBBPA	<LOD			
			ΣTBBP-A	~200-8,000			
			ΣBr	4–17,000			
			ΣBTBPE & DBDPE	Trace			
Leslie et al. (2016)	NLD	European ELV parts		0.2		Conc.'s indicate some legacy contamination of secondary waste stream but at low levels Upper range limit of BDE209 (a candidate POP) in toys cause for concern <0.06–800 µg g^{-1}	
		US/Asian ELV parts		0.3–25,000	*		
		WEEE items		0.5–800	†		
		Shredded car plastic		0.1–11			
		Shredded car & WEEE plastic (mix)		1–280			
		Shredded WEEE plastic		2–330			
		Recycled plastic pellets		0.7–67			
		Insulation/carpet padding		0.001–0.04			
		Office & kitchen products		0.005			
		Plastic toys	ΣPOP-BDE	0.01–33			
Lyu et al. (2015)	CHN, Beijing	Wash basin, litter basket, mat, plastic stool, mop, kettle, PPR pipe, PE pipe, PVC pipe, slippers, luggage & folder	ΣPBDE	5.98 (0.45 - 21.30)		Very low content in all samples	A
	CHN	PVC wastes	ΣPBDE	61.9			

Ref.	Context	Samples	Substance	Mean or range conc. (µg g^{-1}) plastic		Key findings	USMR[#]
			ΣHBCD	18.7		Results indicate contamination from legacy materials as BFRs not believed to be widely used in PS and PVC	
			ΣPBDE	388.0			
Hong-Gang et al. (2016)		PS wastes	ΣHBCD	20.8			
		PS (residual packaging waste)		4.4			
		PP (residual non-packaging waste)		3.0			B
		NSP (residual non-packaging waste)					
	DNK		TBBPA	2.2			B
	CHN, DNK, DEU, NLD		DBPs	8			
			2,4,6-TBP	340			
		ABS (recycled)	TBBPA	26,000	*		
		PS (residual packaging waste)		0.5			
				5.1			
	DNK	EPS (residual packaging waste)		330			
	CHN, DNK, DEU, NLD	PS (virgin)		0.01			
		PS (recycled)		0.76			
		PET (non-packaging waste)		1.3			B
		NSP (non-packaging waste)		3.2			B
		NSP (packaging waste)		0.27			
				0.63			
	DNK	Foil laminated (packaging waste)	ΣHBCD	0.19			
		PP (waste packaging)		(4)			
	DNK	PET (waste packaging)		(1)			
		HDPE (virgin)		(2)			
		LLDPE (virgin)		(1)			
		PP (virgin)		(3)			
		PS (virgin)		(5)			
		PET(G) (recycled)		(2)			
		PET (recycled)	PBDEs (presence only - number of congeners detected in brackets)	(2)		The presence of BFR in multiple samples indicates use of secondary plastics in applications which pose a risk to human health such as food contact materials and toys	
		HDPE (recycled)		(2)			
	CHN, DNK, DEU,	LDPE (recycled)		(3)			
		LLDPE (recycled)		(1)			
Pivnenko et al. (2017)	NLD	PP (recycled)		(2)			

[*] Conc. > RoHS and POP Directive threshold of 1,000 µg g^{-1} ; [†] Conc. > proposed amendment to POP Directive 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); expanded polystyrene (EPS); hexabromocyclododecane (HBCD), 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).

Three studies identified BFRs in products at levels exceeding the RoHS limit and a further three exceeded the limit proposed in the amendment to the POP Directive (European Union, 2019). Of these, Leslie et al. having identified BFRs in end of life vehicle (ELV) car parts is perhaps the least concerning. 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. The sample of recycled acrylonitrile butadiene styrene (ABS) analysed by Pivnenko et al. (2017) (26,000 $\mu g\ g^{-1}$) highlights lack of control over the sources of secondary feedstock, and risks an unbroken chain through which the presence of substances such as BFRs may be sustained in the value chain. However, again the source of the sample is 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. 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. While all the toys purchased in Europe were manufactured in China or Turkey, their presence in such high quantities is noteworthy and 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 2** with data from interviews with stakeholders in the Dutch waste management sector to estimate that 22% 'POP-BDEs' from waste electrical and electronic equipment (WEEE) plastics and 14% POP-BDEs from ELV plastics are recycled into new products in the Netherlands. The assumptions made by Leslie et al. are strongly driven by the opinions of stakeholders working in the Dutch waste management sector and this factor should be taken into account when considering their findings. However, even small quantities of BFRs being re-circulated in this way is a cause for further investigation to ascertain the scale of the potential transgression.

Two other studies identified concentrations of BFRs in plastic products and used them to model exposure to humans and risk to health (**Table 3**). Both studies were based on product

systems in China and reported the main exposure pathway for BFRs to be through inhalation, and in each case, the risk was considered very low and a very small proportion of exposure in the context of other sources of BFRs which mainly result in exposure through being ingested in food.

Table 3: Studies quantifying BFR exposure and risk to human health.

Ref.	Context	Approach	Substance	Receptor	Exposure / risk	Key findings	USMR[#]
Lyu et al. (2015)	CHN, Beijing	Analysis of BFR conc. in products combined with modelling to assess exposure through breathing, dermal contact and oral intake	Daily total hazard exposure PBDE	Adults	295.77, 44.29, 0.00 pg (kg·d)	• Breathing identified as primary exposure pathway	A
				Children	769.55, 40.83, 1.91 pg (kg·d)		
			Non-carcinogenic risk from PBDE	Adults	2.28×10^{-4}	• Non-carcinogenic risk much lower than 1.0, the standard recommended by US EPA	
				Children	5.46×10^{-4}	• The health risk of PBDEs in plastic products were considered acceptable	
Chen et al. (2009)	CHN[a]	Analysis of BFR conc. in products combined with modelling to assess exposure through breathing, dermal contact & oral intake	Daily hazard exposure to BFR	Children (3 month – 14 y)	82.6 to 8,992 pg kg^{-1} pw-d	• Likely small proportion of total daily exposure	

[#] = 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); [a] (south) Guangzhou City. Abbreviations: plastics waste (pw); polybrominated diphenyl ethers (PBDEs); Brominated flame retardants (BFR).

3.2. Phthalates

Phthalates are a group of man-made substances used in a variety of industrial applications, but primarily in plastics (mainly polyvinyl chloride - PVC and cellulose polymers) where they are added to increase flexibility. 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; volatilised 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 signalling in the endocrine system. In animal studies, phthalates have been shown to be carcinogenic endocrine disruptors 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 et al., 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).

In 2019, phthalate production was 6.76 Mt an increase of 21% over the previous five years (4.79% compound annual growth rate - CAGR) (Benjamin et al., 2017). Of this, 80% wt. used in plastics is incorporated in PVC, with the remainder being used in cellulose polymers (Hahladakis et al., 2018; Wang et al., 2011).

The migration of phthalates into foodstuffs has been demonstrated in multiple studies (Hahladakis et al., 2018) and thus they 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 used. However, their presence is near ubiquitous throughout the environment (Gao and Wen, 2016), and the potential harm phthalates may cause to human health has elicited anxiety and confusion amongst some people in society over the extent to which plastics contain them and the level of exposure which people may be subjected to (Carter, 2012; Entine, 2011; Putrich, 2015).

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 4**). In both studies (Pivnenko et al., 2016; Simoneit et al., 2005), phthalates were identified in materials or products where phthalates are not added intentionally, namely non-PVC and non-cellulosic plastics. Contextualising 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 1,000 µg 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 they contained phthalate plasticised PVC. In these examples, the phthalate content would be unsurprising. In a specific example, the ‘roadside trash’ sample analysed by Simoneit et al. contained PVC of unknown origin and it is hence unsurprising that 2,164.7 µg

g^{-1} (0.21%) was identified, given that plasticised PVC may contain between 10 and 70% by weight (wt.) of intentionally added plasticiser.

Table 4: Phthalates identified in secondary plastics.

					Conc. µg g^{-1} plastic			
Ref.	**Geog.**	**Samples**	**Polymer**	**Analyte**	**Mean**[†]	**Min**	**Max**	
		New shopping bags	PE		203			
		'Roadside trash'	[a]		2164.7*			
Simoneit et al.	CHL	'Landfill trash'	[a]		230.9			
(2005)	USA	New shopping bags	PE	DEHP	ND			
			NS		1,894*	832	2,976*	
			NS		416	348	494	
			PP		26	13	95	
			PET		23	6	51	
			HDPE		16	14	21	
		Source segregated plastic waste	PS		11	1	29	
			NS		2	2	3	
			NS		215	28	640	
			PP		150	145	157	
			LDPE		103	99	139	
			HDPE		51	49	55	
			HDPE		46	43	48	
			NS		30	27	35	
			PP		19	15	24	
		Reprocessed household plastics	HDPE		7	5	10	
			PET		2	0	6	
			NS		567	481	1,028*	
			NS		522	336	1,071*	
			LDPE		186	99	570	
			NS		144	68	282	
			PS		103	94	110	
			PS		66	55	97	
			NS		52	14	272	
			PET		49	33	88	
			NS		46	46	47	
			PP		33	32	34	
			PP		6	5	12	
			HDPE		4	2	10	
Pivnenko et al. (2016)	CHN, DNK, DEU, NLD	Residual plastic waste	PET	Σ Phthalate	4	1	8	C

[a] Composition: PE 17.3%, PET 29.7%, PVC 39.3%, PS 2.9%, unidentified plastics 10.8%; [b] samples ground into 20 samples: residual (n=13) and source segregated (n=7); virgin plastics (n=8), recycled plastics from households (n=9); recycled plastics from post-industrial (n=11); * Conc. > US Code (2008) and Consumer Product Safety Commission (2017) threshold of 1,000 µg g^{-1} in toys; [#] uncertainty, strength of knowledge and methodological robustness (USMR) assessed qualitatively. It is assumed that there are no significant concerns unless marked as: C = high conc. were in unidentified (not specified) samples. Abbreviations: Di(ethylhexyl) phthalate (DEHP); not specified (NS); low density polyethylene (LDPE); high density polyethylene (HDPE); polypropylene (PP); polystyrene (PS); polyethylene terephthalate (PET); geographical context (Geog.); concentration (conc.)

Nonetheless, the presence of phthalates in such a wide range of samples (**Table 4**), 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 because of their introduction during bottling.

Whereas the assessment of harm to human health carried out by Keresztes et al. (2013) was based on bottled waste 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 5**). 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.

Table 5: Phthalate exposure and risk to human health in Denmark; after Lee et al. (2014).

Approach	Substance	Receptor	Exposure / risk	USMR[#]
Mass flow used to inform assessment of childhood phthalate exposure using stochastic modelled scenarios including case study of two-year old children living in Denmark	DEHP, DBP, BBP	Population	4 to 18% re-enters product cycle through recycling of which 2 to 12% re-circulated inside Europe.	
	DBP	Children	Increasing recycled content in paper and PET food packaging could increase exposure by 0.116–0.355 μg kg^{-1} bw d (18% of exposure for two-year-olds living in Denmark)	E

[#] = Uncertainty, strength of knowledge and methodological robustness (USMR) assessed qualitatively. It is assumed that there are no significant concerns unless marked as: E = a probabilistic approach was taken to model phthalate exposure. An assumption was made that 'micro-contaminants' would not be regulated. While this may be a reality, especially with goods from the Far East, it is speculative. Further, migration of phthalates was not modelled, reducing the strength of the findings in this article. Abbreviations: Dibutyl phthalate (DBP); Benzyl butyl phthalate (BBP); Di(ethylhexyl) phthalate (DEHP); body weight (bw).

While the studies summarised in **Table 4** and **Table 5** 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 (PTE)

Some elements represent a potential hazard to human health due to their toxicity at relatively low concentrations. Lead, chromium, 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 S 7**).

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 stabilisers; 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) indicate that some PTEs may be passed along the value chain as a legacy from their previous use (**Table 6**). In both studies, the concentrations were reported to be 'low' for all elements; however, as Whitt et al. indicate, 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. Furthermore, 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 confirm this probability, but they were not carried out in either of the two studies, meaning that the potential hazard exposure from the concentrations identified was not determined. Nonetheless, Eriksen et al. (2018) points out that as 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.

Table 6: Elements including potentially toxic elements (PTE) identified in secondary plastics.

Ref.	Context	Samples	Metal	Conc. µg g^{-1} plastic: Mean	Standard error	Key findings	USMR[#]
Eriksen et al. (2018)	DNK	Samples of PET, PE, PP, PS (n=82), from reprocessed HH plastic waste, HH plastic waste (are), pre-consumer waste, and virgin plastic	Al	269		• All elements detected in all samples at levels below RoHS Regulations thresholds • Levels were higher in waste samples compared to non-waste samples, indicating accumulation as a consequence of recycling • Differences in metal concentrations associated with material chemistry rather than contamination with the exception of Mn	F
			As	0.11			
			Cd	2.52			
			Co	2.47			
			Cr	6.59			
			Cu	7.71			
			Fe	158			
			Hg	1.26			
			Li	0.70			
			Mn	2.63			
			Ni	1.84			
			Pb	3.39			
			Sb	34.30			
			Ti	2,090			
			Zn	35.10			
Whitt et al. (2012)	USA, CHN	Rigid RPET food contact packaging containers	Cd	8.82	0.84	• None of the samples exceeded the State of California Health and Safety Code incidental limit of 100 µg g^{-1} (total 'heavy metals') • Sb is likely present as a residual catalyst used to produce thermoformed PET whereas other elements are likely contaminants from previous use	G
			Cr	6.76	0.59		
			Ni	9.43	0.90		
			Pb	0.15	0.02		
			Sb	8.30	0.48		
		RPET food contact packaging film	Cd	18.93	1.28		
			Cr	13.63	0.96		
			Ni	19.89	1.32		
			Pb	0.16	0.02		
			Sb	3.62	1.77		

[#] = Uncertainty, strength of knowledge and methodological robustness (USMR) assessed qualitatively. It is assumed that there are no significant concerns unless marked as: F = link with accumulation inferred, but not statistically correlated; G = potential to migrate into food contained in packaging was discussed, but no migration testing performed. Abbreviations household (HH); restrictions on hazardous substances (RoHS)
polypropylene (PP); polystyrene (PS); polyethylene terephthalate (PET); polyethylene (PE); recycled polyethylene terephthalate (RPET);

3.4. Other volatile organic compounds (VOC)

The term 'volatile organic compounds' (VOCs) is a coverall for a wide range of substances that evaporate at room temperature. VOCs can be divided into three broad sub-groups according to volatility as shown in **Table 7**.

Table 7: Classification of volatile organic compounds (VOCs); after United States Environmental Protection Agency (2017).

Description	Abbreviation	Boiling point range (°C)	Example compounds

Very volatile (gaseous) organic compounds	VVOC	<0 to 50-100	Propane, butane, methyl chloride
Volatile organic compounds	VOC	50-100 to 240-260	Formaldehyde, d-Limonene, toluene, acetone, ethanol (ethyl alcohol) 2-propanol (isopropyl alcohol), hexanal
Semi volatile organic compounds	SVOC	240-260 to 380-400	Pesticides (DDT, chlordane, plasticizers (phthalates), fire retardants (PCBs, PBB))

Abbreviations: polychlorinated biphenyls (PCB); dichlorodiphenyltrichloroethane (DDT); polybrominated biphenyl (PBB).

VOCs occur throughout the natural environment and volatility is not necessarily an indicator of potential hazardousness. However, some VOCs are carcinogenic and many have been found to irritate lungs, exacerbate allergies and damage the central nervous system (Kwon et al., 2018; United States Environmental Protection Agency, 2017).

In plastics, VOCs may be of concern because their volatility increases the likelihood of migration to the surface via thermodynamic equilibrium, and subsequent release into the atmosphere. VOCs may exist already in plastics as intentionally added substances, arise as contaminants picked up during the use phase or through waste management practices, or as a result of transformation through oxidation or degradation of these additives or of the polymer itself (Skjevrak et al., 2003). Therefore, characterisation of VOCs in plastics or in the emissions from heated or combusted plastics can provide an indication of the origin of the plastic material and the products it contained in its previous application.

In this review, three examples of VOC concentrations in air, which indicate cross contamination of plastics, were identified by Tsai et al. (2009) (**Table 8**). All were in Taiwan and no indication of the prevalence across other similar facilities is given. In these cases, the lack of control over the source of input materials is indicative that VOCs, and also other unknown substances, may be inherited from previous use or processing. Additionally, the atmospheric emissions themselves cannot be adequately controlled if the compounds being emitted cannot be anticipated. For instance, 1-3 butadiene, a carcinogen Sielken and Valdez-Flores (2015), was observed to be present in the air inside the PVC plant at levels 7–17 times greater than indoor air reference levels (IARL) proposed by Health Canada (2018). In this case, the risk manager of the plant would not be able to anticipate and therefore calculate the exposure to the workforce without constant, costly air emissions monitoring.

Table 8: Concentrations of volatile organic compounds (VOCs) observed in air samples at recycled plastic extrusion facilities in Taiwan, China. Selected examples are inconsistent with virgin production for each specified plastic after (Tsai et al., 2009).

Plant type	Substance	Conc. in air (µg m^{-3})	IARL (µgm^{-3})[a]	Key findings	USMR[#]
	Acrylonitrile	64-67			
	1,3-butadiene	12-29	1.7		
PVC plant	Styrene	168-480	850	Likely cross-cont. ABS waste	
PP plant		30-36			
PE plant	Cl-comp	11-54		Likely cross-cont. PVC waste	H

[a] Indoor air reference levels set by Health Canada (2018); [#] Uncertainty, strength of knowledge and methodological robustness (USMR) assessed qualitatively. It is assumed that there are no significant concerns unless marked as: H = no significant concerns, however the intended use of products made from the plastics discussed was not stated so the impact of content of inconsistent materials on health cannot be determined.
polyethylene (PE) polypropylene (PP); polyvinyl chloride (PVC).

As the study investigated air emissions of VOCs generated during thermal processing of plastics, no indication is provided by Tsai et al. (2009) of the risk to human health through normal use of the plastics that have been contaminated by other polymers and their constituent additives.

3.5. Risk characterisation for legacy materials in secondary plastics

The qualitative risk characterisation for legacy materials in secondary plastics found generally low to low/medium harm potential across each hazard group (**Table 9**). The greatest risk identified was through VOC emissions generated in plastics reprocessing plants at the extrusion unit operations, where plastics such as PVC and ABS are allowed to cross-contaminate other plastic types. 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.

The identification of BFRs also resulted in a low/medium risk score. Several secondary plastics were identified with concentrations that exceeded POP Directive limits, but data quality was impeded by lack of detail on sampling choices.

Both phthalates and PTEs were scored as a low potential risk. However, in the case of both hazard groups there is an inference that plastics recyclers in LIMICs and HICs are not adequately controlling supply chains to reduce the risk of contamination of products made from secondary plastic materials. Further research is recommended to ascertain the prevalence and quantities of legacy contamination across all four hazard groups identified in this section.

Table 9: Risk characterisation summary for legacy substances in 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
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; Guzzonato et al., 2017; Leslie et al., 2016; Lyu et al., 2015; Pivnenko et al., 2017). • Five samples of toys and two samples of food contact items had levels of BFRs that exceed revised POP Directive 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 whether: 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 (Eriksen et al., 2018; Whitt et al., 2012) 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 (2006) 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 recycling 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

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).

4. 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 usually (not always) comminuted, before being melted under pressure in an extruder which forces the then molten material through a die for direct product production or palletisation. The heat, between 200-300 °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 into secondary products. If unabated, it is inevitable that workers and residents in the vicinity will be exposed to these vapours and gasses, many of which can be detrimental to their health. In addition to the exposure from inhalation, particle and vapours may be carried away from extrusion plants on the wind and be deposited into surrounding environmental media such as soils, dust and sediments (**Figure 4**).

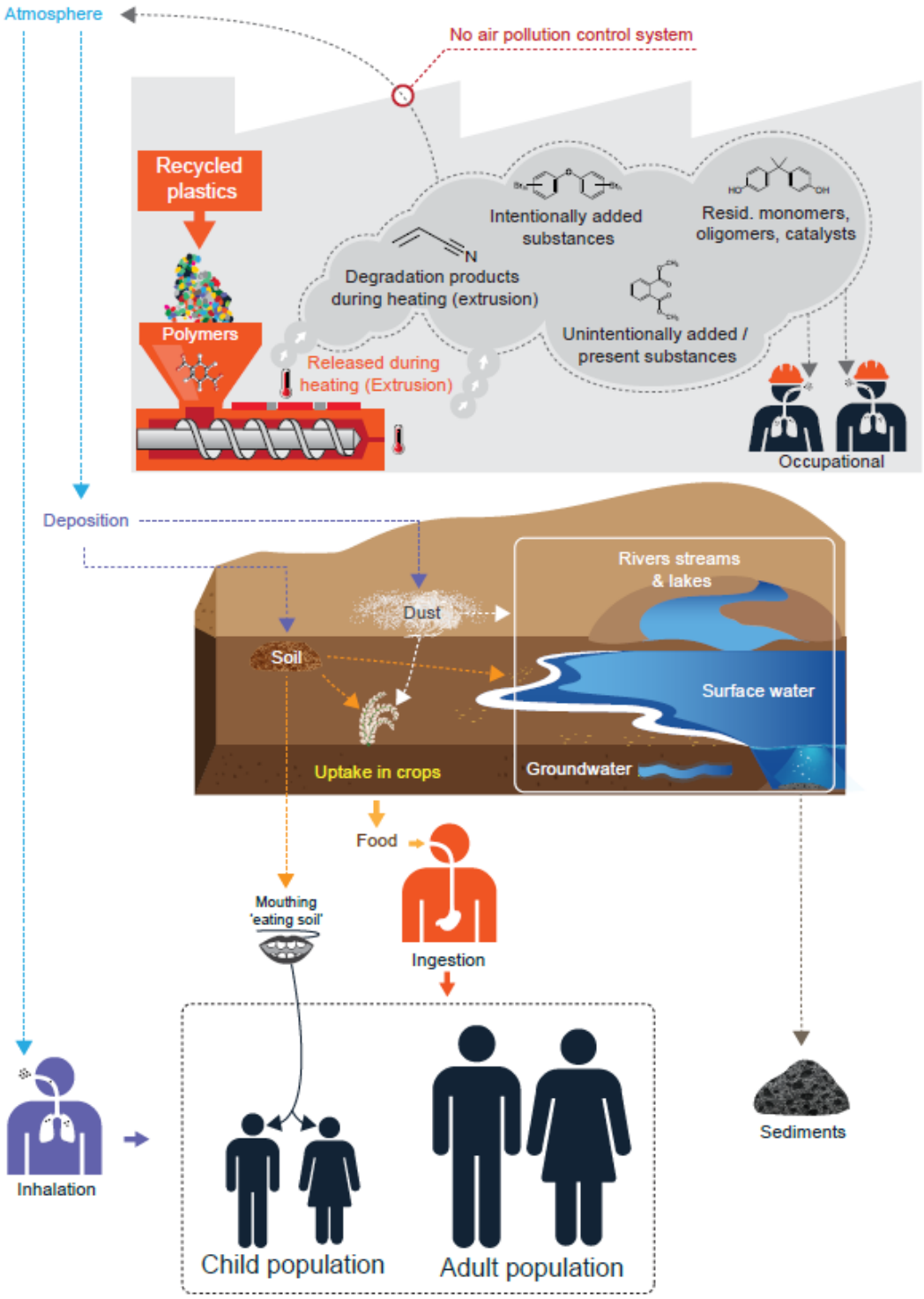


Figure 4: Hazard exposure conceptual model (source–pathway–receptor) associated with extrusion of plastic waste.

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 (Health and Safety Executive, 2013; Unwin et al., 2012). 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 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.

The following section reviews the evidence for exposure to potentially hazardous substances released during extrusion, focusing on studies which have quantified atmospheric emissions from extrusion practices, and which have analysed soils and sediments nearby to plastics recycling facilities.

4.1. Brominated flame retardants (BFR)

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. In this review, two papers by Tang et al. (2014) and (2015) determined concentrations of BFRs in soils, sediments and road-dust in a district of China where significant plastics recycling has taken place over recent decades (**Table 10**). Most samples showed much higher concentrations compared to samples collected from other parts of China and Asia where no plastics recycling takes place (**Table 11**). Neither paper 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 significant plastics and/or e-waste recycling activities take place.

Table 10: BFRs identified in soil, sediment and dust samples near to plastics recycling facilities as well as human hair samples from residents and plastics recycling workers.

					Conc. ng g^{-1} (dry wt.)		ΣTEQ pg TEQ g^{-1}			
Ref.	**Context**	**Samples**		**Substance**	**Mean**	**Range**	**Mean**	**Range**	**Key findings**	**USMR[#]**
			Zhaogezhuang (n=40)		690	(1.25−3,673)	10.0	(0.01−54.1)		
		Soil	Daliu (n = 22)	ΣPBDE	437	(3.11−5,504)	6.20	(0.04−78.5)		
			Xiaobaihe River (n=10)		190	(18.2−1,435)	2.71	(0.22−20.9)		
			Renwen Canal (n=6)		2,841	(210−9,889)	41.23	(2.94−144)		
		Sediment	Yincun Ditch (n=9)	ΣPBDE	2,314	(276−7,317)	33.1	(3.97−107)	• Plastics recycling operations are a significant source of PBDE in soils and sediments in this area of China	I
			Children (n=11);		89.6	(1.50−418)	1.27	(0.005−5.72)	• No significant gender differences in concentrations in hair samples	
			Young (n=16);		133	(2.14−861)	1.85	(0.03−12.03)		
Tang et al. (2014)	Hebei, CHN[a]	Hair	Middle-aged (n=18)	ΣPBDE	108	(6.06−587)	1.51	(0.09−8.34)	• Higher conc. in young group could be a result of occupational exposure	J
Tang et al. (2015)	Hebei, CHN[a]	Road dust (n=20) Residential dust (n=11)		Σ_{21}PBDE	1,541	(±2,348)	23.1	(±36.1)	• Mean hazard quotient for adults 0.00577 indicating low overall health risk	

[a] = Wen'an County, northeast Hebei Province, China; main cottage industry plastics recycling area in northern China reported to house several hundred plastic recycling facilities for over 30 years; [#] = Uncertainty, strength of knowledge and methodological robustness (USMR) assessed qualitatively. It is assumed that there are no significant concerns unless marked as: I = no significant concerns, however the plastics recycling activities carried out in the area are not clearly specified, so there is ambiguity as to whether they are related to open burning or residues or extrusion emissions; J = aggregation of samples by age rather than exposure means it is not possible to distinguish between them. Abbreviations: polybrominated diphenyl ethers (PBDEs); concentration (conc.); toxic equivalency (TEQ).

Table 11: PBDE concentrations (ng g^{-1} d) in sediment samples at various locations in China and other Asian countries; adapted from Tang et al. (2014).

			BDE 209		ΣPBDEs	
Location	**Sampling dates**	**Sampling surroundings**	**Mean**	**Range**	**Mean**	**Range**
Nanyang River, Guiyu, China	Dec 2004	Near to e-waste open burning and dumping site	35.9	16.9−62.2	9,357	4,434−16,088
Lianjiang River, Guiyu, China	Dec 2004	Near living location, e-waste recycling region	30	15−57	156	52−365
Maozhou River, Shenzhen, China	Jan 2010	Surface sediments from urban areas	449	16.5−2,673	879	26.6−4,885
Pearl River, China	Sep 2002	Urbanised areas	890	26.3−3,575	903	27.7−3,587
Yangtze River Delta, China	Apr 2002	Intertidal zone of the YRD		0.16−94.6		0.16−95.1
Tributaries of Haihe River, China	2009	14 principal tributaries of Haihe River	0.02	ND−0.13[b]	0.80	0.06−2.10
Fuhe River, China	2007−2008	Major inflowing river of Baiyangdian Lake	103		118	
Baiyangdian Lake, China	2007−2008	Largest natural freshwater body in North China Plain	10.4			14.7
Asian canals or rivers	2000−2010	Capital or major cities of eight tropical Asian countries or districts		0.54−1,670		0.83−3,140
Tokyo Bay, Japan	2008	Sewage and urban run-off		2.68−232		34.1−465

[b] ND, concentration lower than detection limit. Abbreviations: polybrominated diphenyl ethers (PBDEs); brominated diphenyl ethers (BDEs); not detected (ND)

Tang et al. (2014) contextualised 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 recycling 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 (2015) in soils, sediments and dust may have been transported there in the atmosphere, having been volatilised during re-extrusion of e-waste or end-of-life vehicle (ELV) parts being several decades old, or given that many BFRs are still on the market in South East Asia, from re-extrusion of relatively new materials. However, given the high boiling points of this class of compounds (~250 to 450°C), it seems likely that open burning of unwanted plastic residues may also be a significant, and possibly greater source.

BFR degradation mechanisms in the environment are only partly understood and the subject of continuing research (Lassen et al., 2014). Tetrabromobisphenol A (TBBPA) has been shown to degrade both aerobically and anaerobically (Lee et al., 2011), into mainly bisphenol A, but also tetrabromobisphenol-A-bis(methyl ether) which has a greater potential to bioaccumulate in comparison to TBBPA itself. Other research (European Chemicals Bureau, 2008) indicates full mineralisation of TBBPA in soils with a half-life of six months. Photolysis is thought to be an important mechanism for debromination of PBDEs, particularly the more brominated homologues (Schenker et al., 2008).

Deca-BDE has been reported to have a typical half-life of more than one year in soils, however some studies shown no degradation at all in anaerobic sediments 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. BFRs pose a risk to the surrounding population as they may be taken up by food crops and directly ingested by children who are inherently more sensitive to exposure.

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 are likely to be uncomprehensive in these countries, emissions of BFRs may

result in significant harm to both extrusion workers, but also to residents living in the vicinity of poorly managed plastics recycling facilities worldwide.

4.2. Phthalates

Three studies reported phthalate emissions from extrusion of secondary plastics (**Table 12**). The studies use non-comparable variables and units, and are therefore described independently.

Table 12: 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 • Exhaust gasses not controlled at plastics extrusion facilities in region • Steps should be taken to provide workers with respiratory protective equipment	K
						Gas phase		938.8	SD 56.1		
				Styrene-butadiene copolymer (K-resin) plant		Particle phase		279.7	SD 26.6		
						Gas phase		624.7	SD 47.5		
			Outside	PC-ABS plant		Particle phase		94.7	SD 12.0		
						Gas phase		266.2	SD 25.2		
				Styrene-butadiene copolymer (K-resin) plant		Particle phase		68.9	SD 11.7		
						Gas phase		144.6	SD 14.9		
			Outside	Reference courtyard (20 km distant)		Particle phase		40.9	SD 7.3		
						Gas phase		27.6	SD 4.1		
Yamashita et al. (2009)	JPN	Atmospheric emissions sampling in laboratory	Virgin [c] LDPE, PP, PS					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	DMP		µg g^{-1d}	2.94	±0.67		
					DEP			2.47	±0.65		
			Mix PE, PP, PS recycled pellet	Heated in N_2	DMP			3.14	±1.0		
					DEP			1.90	±0.30		
Wang et al. (2011) [b]	Hunan, CHN	Field sampling of environmental media	Well water	Exposed	DEHP		µg g^{-1}	14.2	(0.32–87.70)		L
				Reference				0.79	(0.28–2.27)		
			Pond water	Exposed				135.68	(0.31–429.89)		
				Reference				0.37	(0.23–0.47)		
			Industry wastewater	Exposed				42.43[a]	(0.36–161.86)		
				Reference			µg L^{-1}	n/a			
			Agricultural soil	Exposed				13.07	(0.85–37.23)		
				Reference			µg g^{-1}	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 recycling area. Not correlated with particular plastics recycling operation. Cause of phthalate release not determined. Could be result of open burning of residues; comminution; agitation; or extrusion. Abbreviations: dimethyl phthalate (DMP); diethyl phthalate (DEP); di-2-ethylhexyl phthalate (DEHP); polycarbonate acrylonitrile butadiene styrene (PC-ABS); concentration (conc.).

The study by Huang et al. (2013) of phthalate concentrations in indoor and outdoor air at recycled ABS and K-resin plants in China highlighted concern over exposure to workers. However, the levels observed were far below the mean long term Workplace Exposure Limits (WEL) over eight hours of 5,000,000 ng m^{-3} recommended by the Health and Safety Executive (2020a) in the UK.

Yamashita et al. (2009) observed phthalate emissions from secondary and virgin plastics where they had not been intentionally added. The emissions were identified alongside chlorinated compounds, which Yamashita et al. suggest are a result of contamination of each polymer with PVC, which is commonly plasticised with phthalates. The results, expressed per gram of plastic, were not compared with other previous evidence in the study by Yamashita et al. and no other studies were revealed in this review, which means the prevalence of this phenomenon has not been estimated.

In the third study, by Wang et al. (2011), water and soil samples from a plastics recycling region in China were analysed for phthalate concentration and compared with reference samples. The plastics recycling area showed concentrations orders of magnitude greater than the reference areas indicating that the plastics recycling operations were a significant source. To add context, Wang et al. (2011) also analysed blood of occupationally exposed recycling workers, concluding that working in the plastics recycling 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 13**).

Table 13: Phthalate levels observed in urine and blood of occupationally and environmentally exposed people.

Ref.	Year	Context	Approach	Substances	Exposure/risk	Key findings	USMR[#]
Petrovičová et al. (2016)	2016	Nitra Region, SVK	Urine analysed for workers in: • Waste collection (n=45) • Plastic extrusion (n=35) Students (control) (n=49)	MEHP; MEHHP; MEOHP; ∑DEHP; MiBP; MnBP	Significantly (p<0.02) higher conc. in urine from plastics extrusion workers compared to waste management workers and control group		M
Wang et al. (2011)	2011	Hunan, CHN	Blood samples analysed from 181 plastics recycling workers and 160 gender-age matched farmers (samples collected in August 2008)	8-hydroxy-2′-deoxyguanosie (8-OHdG)	Significantly (p<0.01) higher in plastics recycling workers compared to reference: Mdn: 340.37 μmol mol^{-1} creatinine (Q1 222.39; Q3 511.01)	Correcting for sex, working in plastics recycling was a significant independent predictor of higher urinary 8-OHdG (OR=2.323, p<0.01) for male workers but not female workers. Male workers may experience oxidative damage to DNA as a consequence of phthalate exposure.	N
				Serum malondialdehyde (MDA)	Significantly (p<0.01) higher in plastics recycling workers compared to reference: Mdn: 3.80 nmol ml^{-1} (Q1 2.87; Q3 4.72)		
				Serum superoxide dismutase (SOD) activity	Significantly (p<0.01) lower in plastics recycling workers compared to reference: Mdn: 112.15 U ml^{-1} (Q1 100.87; Q3 124.62)		

[#] = Uncertainty, strength of knowledge and methodological robustness (USMR) assessed qualitatively. It is assumed that there are no significant concerns unless marked as: M = history of workers and the types of plastic being processed unclear; N = samples collected from historical plastics recycling area. Not correlated with particular plastics recycling operation. Cause of phthalate release not determined. Could be result of open burning of residues; comminution; agitation; or extrusion. Abbreviations: monoethylhexyl phthalate (MEHP); Mono(2-ethylhexyl) phthalate (MEHHP); mono(2-ethyl-5-oxohexyl) phthalate (MEOHP); di(ethylhexyl) phthalate (DEHP); mono-n-butyl phthalate (MnBP) and mono-isobutyl phthalate (MiBP); odds ratio (OR).

One other study by Petrovičová et al. (2016) detailed in **Table 13,** analysed the urine of Slovakian workers in the plastics extrusion sector, finding significantly (p<0.02) higher concentrations 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 are from recycled feedstock, therefore there is little relevant conclusive evidence which adds to this review.

While the studies summarised in **Table 13** indicate some cause for concern, the risk to the workers was not modelled. 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 significant cause of harm to workers or residents in the areas surrounding these plants.

4.3. Other volatile organic compounds (VOC)

Volatile organic compounds (VOCs) are produced during plastics extrusion due to the length of thermal exposure, and the interactions between the polymer and the various additives, polymerisation residues, and unintentionally present substances. VOCs readily evaporate at room temperature, so when plastic is extruded at between 150 and 300°C, any VOCs present are readily released into the surrounding atmosphere (Hahladakis et al., 2018).

This review identified three studies at 11 plastics recycling 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 14**.

Table 14: Comparison of average total VOC concentration (μg m^{-3}) observed at recycled plastics extrusion facilities.

	Tsai et al. (2009) Taiwan, CHN		**He et al. (2015) CHN**			**Huang et al. (2013) Guangdong, CHN**			
Plastic extruded	**Inside**	**Temp. (°C)**	**Inside**	**Outside**	**Temp. (°C)**	**Inside**	**Outside**	**Ref.**	**Temp. (°C)**
ABS			1,000,000	1,800	200-300				
PC			990	600	100-150				
PC-ABS						3,722.5	2,784.7	726	230-300
PS			470,000	3,900	200-260				
PE	717-3,192	260–270	2,800	1,100	150-250				
PP	1,625-6,050	230–250	59,000	1,200	150-250				

	Tsai et al. (2009) Taiwan, CHN		He et al. (2015) CHN			Huang et al. (2013) Guangdong, CHN			
Plastic extruded	**Inside**	**Temp. (°C)**	**Inside**	**Outside**	**Temp. (°C)**	**Inside**	**Outside**	**Ref.**	**Temp. (°C)**
PVC	7,892-28,864	180	25,000	1,900	150-200				
PA			26,000	2,100	200-230				
K-Resin						3,223.20	2,767.9	726	200-230

Abbreviations: polycarbonate/acrylonitrile-butadiene-styrene (PC-ABS), styrene-butadiene copolymer (K-resin); polyamide (PA); acrylonitrile-butadiene-styrene (ABS); polycarbonate (PC); polypropylene (PP); polyvinyl chloride (PVC); polystyrene (PS); polyethylene (PE); temperature (temp.)

Levels of VOCs in one of the ABS plants and one of the PS plants studied by He et al. (2015) both showed very large emissions in comparison to all other studied facilities. In both cases, styrene dominated the emission profile (data not shown), representing 63% (ABS 630,000 μg m^{-3}) and 65% (PS 310,000 μg m^{-3}) of the total VOCs emitted. Total VOC emissions at the PE plants investigated by He et al. (2015) and Tsai et al. (2009) reported low concentrations within a similar range in comparison to other polymers extruded, except for the PC plant, where the lowest concentration was observed.

He et al. (2015) extrapolated field sampling to model long term risk from VOC exposure for workers inside the plants and for residents living nearby, finding chronic acute risks to health workers inside the ABS and PS plants (**Table 15**). The modelling indicated no significant risk to workers or residents from the PP, PE and PC plants, however it inferred that emissions of VOCs from PS, PA, ABS and PVC plants would lead to a risk of cancer to residents over their lifetimes if emissions were not mitigated.

Table 15: Summary of health risk assessment carried out by He et al. (2015) for workers in plastics extrusion plants in China and residents living nearby.

Plastic extruded	**Substance**	**Receptor**	**Exposure / risk**
	Benzene, toluene, ethylbenzene, styrene, methylene chloride and trichloroethylene		Major contributors towards chronic non-carcinogenic health effects in these workshops.
PS, PA, ABS, PVC	Acrylonitrile, styrene, ethylbenzene and 1,2-dichloromethane verified as Group 2B		Major contributors to cancer risks, like tumour of the lungs, liver, kidneys, and brain via inhalation exposure.
PS ABS		Workers	Acute, chronic risks to health.
PS, PA, ABS, PVC			Risk of cancer over their lifetimes, however the non-cancer risks were unlikely to affect residents.
PE, PP, PC	Total VOC	Residents	Not found to pose significant risk to health of workers or residents.

Abbreviations: volatile organic compounds (VOC); polypropylene (PP); polystyrene (PS); polycarbonate (PC); polyethylene (PE); acrylonitrile-butadiene-styrene (ABS); polyamide (PA); polyvinyl chloride (PVC);

Field studies of emissions from plastics extrusion plants are complemented by Yamashita et al. (2009) who heated samples of virgin and recycled plastics and observed VOC emissions in the laboratory (**Table 16**). The study 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. 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.

Table 16: Emissions of volatile organic compounds (VOCs) from combustion of plastics in different atmospheric compositions; after Yamashita et al. (2009).

				Conc. µg m^{-3}
Sampling	**Temp.**	**Atmosphere**	**Substance**	**Mean**
		Air		150
Virgin† LDPE		N_2		100
Virgin† PP		Air / N_2		450
Virgin† PS		Air / N_2		350
		Air	Total VOC	550
Recycled mixed PE, PP, PS pellet		N_2	(in toluene equivalent)	1,200
			Aliphatic HC	30
			Aldehyde	15
			Ketones	10
			Carboxylic acid	10
LDPE	200°C		Esters	0
			Aliphatic HC	240
			Aldehyde	180
			Ketones	120
			Carboxylic acid	90
LDPE	250°C	Air	Esters	60

Abbreviations: volatile organic compounds (VOC); Low density polyethylene (LDPE); polypropylene (PP); polystyrene (PS); polyethylene (PE); hydrocarbons (HC).

Yamashita et al. (2009) also investigated the effect of VOC production on increasing operating temperature of the low-density polyethylene (LDPE), finding a considerable increase in VOC production at 250°C compared to 200°C. 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.

According to Huang et al. (2013), He et al. (2015) and Tsai et al. (2009), control measures to protect workers and nearby residents from exposure to VOCs are seldom implemented in plastics recycling facilities in China; and it is conceivable that this is also the case throughout many other LIMICs. Given the acute chronic and carcinogenic risk to workers in ABS and PS plants, and the carcinogenic risk to workers and residents for the PS, PA, ABS and PVC

extrusion plants, there is an urgent need to carry out more research to widen the evidence base for these practices across LIMICs.

One final noteworthy data-point is that reported by Unwin et al. (2012) who sampled air at ten UK plastics extrusion plants. While the majority of the compounds were PAHs, at one of the plants which extruded PVC, the study also detected low levels of vinyl chloride monomer; 51.78 (0.02 ppm), or 0.7% of the UK HSE workplace exposure limit. The study was intended to assess the efficacy of the HSE's guidance on controlling exposure to fumes in plastics extrusion facilities (Health and Safety Executive, 2013) and confirmed the efficacy across all ten plants including the aforementioned PVC plant. Whereas a series of measures had been put in place to control occupational exposure to fumes from extrusion, local exhaust ventilation and forced mechanical ventilation and dilution were noted at all of the plants.

4.4. Polycyclic aromatic hydrocarbons (PAHs)

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 17**). Inside both plants, levels of PAH with a toxic equivalency greater than 1ng m^{-3} BaP_{eq} were observed in both the gas and particle phase, indicating a significant 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 appeared to not be provided with respiratory protection equipment as the last line of defence in the hierarchy of risk control (Hughes and Ferrett, 2016).

Table 17: Atmospheric PAH concentration observed at recycling plants in China.

						Conc. (ng m^{-3})		Toxic equivalency: Total BaPeq incl. BaP (ng m^{-3})	
Ref.	**Year**	**Context**	**Samples**		**Substance**	**Mean**	**SD**	**Mean**	**SD**
Huang et al. (2013)	2013	Guangdong, CHN	Inside	PC-ABS plant	TPAH (particle)	24.5	8.0	1.1 *	0.3
					TPAH (gas)	872.5	102.9	2.3 *	0.3
				SBC (K-resin) plant	TPAH (particle)	49.5	9.8	1.1 *	0.3
					TPAH (gas)	1,206.5	135.3	2.3 *	0.2
			Outside	PC-ABS plant	TPAH (particle)	9.6	1.7	0.09	0.02
					TPAH (gas)	475.4	51.1	0.7	0.1
				SBC (K-resin) plant	TPAH (particle)	9.0	2.2	0.07	0.01
					TPAH (gas)	753.6	109.0	1.0 *	0.2
			Outside	Reference courtyard (20 km distant)	TPAH (particle)	3.8	0.9	0.00	0.00
					TPAH (gas)	171.4	32.4	0.2	0.04
Unwin et al. (2012)	2013	GBR	Inside	PVC	Naphthalene	<1			
					Other PAHs	<1			
				PVC	Naphthalene	100			
					Other PAHs	<10			
				PVC	Naphthalene	<60			
					Other PAHs	<20			
				PVC alloy	Formaldehyde	2,000			
					Naphthalene	<20			
					Other PAHs	<20			
				PE&PP	Formaldehyde	3,400			
					Naphthalene	<1			
					Other PAHs	<1			
				PE	Formaldehyde	1,700			
					Naphthalene	<1			
					Other PAHs	<1			
				PE, PP, & PS	Formaldehyde	7,200			
					Naphthalene	<1			
					Other PAHs	<1			
				PET	Naphthalene	<1			
					Other PAHs	<1			
				ABS	Naphthalene	<1			
					Other PAHs	<1			
				EPS	Formaldehyde	9,000			
					Naphthalene	<50			
					Other PAHs	<10			

* = Toxic equivalency >1 ng m^{-3}; # = uncertainty, strength of knowledge and methodological robustness (USMR) assessed qualitatively. It is assumed that there are no significant concerns unless marked as: K = two highly specific plastic waste extrusion plants were studied with different operating temperatures which could influence results: PC-ABS: 230-300C; SBC: 200-230C; key findings: PAH concentrations significantly higher inside plants; exhaust gasses not controlled at plastics extrusion facilities in region; steps should be taken to urgently provide workers with respiratory protective equipment. Abbreviations: styrene-butadiene copolymer (SBC); polypropylene (PP); polystyrene (PS); expanded polystyrene (EPS); polyethylene terephthalate (PET); polyethylene (PE); acrylonitrile-butadiene-styrene (ABS); polycarbonate/acrylonitrile-butadiene-styrene (PC-ABS); styrene-butadiene copolymer (K-resin); polyvinyl chloride (PVC); (benzo(a)pyrene equivalent (BaP_{eq}); polycyclic aromatic hydrocarbons (PAH); Total polycyclic aromatic hydrocarbon (TPAH); standard deviation (SD).

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 these plants. 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 from the study 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) observed PAH emissions at 10 plastics extrusion plants in the UK. The study was intended to assess the efficacy of HSE guidance for plastics extrusion operators (Health and Safety Executive, 2013). Overwhelmingly, the emissions of PAHs across all of the plants were very low with none exceeding 0.4% of HSE workplace exposure limits; confirming the efficacy of local exhaust ventilation, general forced air dilution, ventilation and a series of other measures specified in the guidance.

4.5. Risk characterisation for extrusion of secondary plastics

The calculated risk from extrusion of secondary plastics was medium/high in all hazard categories (**Table 18**). HICs were not assessed, as it is assumed that they generally have measures in place to control emissions from extrusion; whereas in LIMICs, it has been reported that emission controls are often not implemented and the respiratory protective equipment is not provided to workers.

Clearly the extrusion of some polymers will result in an emission profile which is more hazardous in comparison to others, but consistently, PS, PA, ABS and PVC all featured as having a greater likelihood of producing more hazardous emissions. It happens that PC-ABS and K-resin were captured in the search in the present study, however although the quantity of production (and therefore reprocessing) could not be verified in this study, it is certainly less than the polyolefins, PET, PS and PVC.

According to the risk assessment (**Table 18**), plastics recycling workers are generally at as much risk of exposure to emissions as residents whose non-carcinogenic risk from VOC exposure nearby ABS and PVC plants was calculated to be very high (He et al., 2015).

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 recycling areas in China. However, whether these arose from open burning, extrusion and/or abrasion can only be speculative with the low level of available evidence.

Table 18: Risk characterisation summary for 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
BFR	Atmosphere/ inhalation	Plastics recycling 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 recycling 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 recycling 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 recycling workers	CHN	• Total VOCs in PS and ABS plants (He et al., 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 (He et al., 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 recycling 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 (Geog.); waste electrical and electronic equipment (WEEE); end of life vehicle (ELVs); brominated flame retardants (BFR); respiratory protective equipment (RPE); low income and middle income countries (LIMICs); polycarbonate/acrylonitrile-butadiene-styrene (PC-ABS); styrene-butadiene copolymer (K-Resin); volatile organic compounds (VOCs); polystyrene (PS); acrylonitrile-butadiene-styrene (ABS); polyamide (PA); polyvinyl chloride (PVC); benzo(a)pyrene equivalent (BaPeq).

5. Conclusions

The attention placed on circular economy principles and practice in recent decades has incidentally supported 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 on a global systematic review of evidence that indicates harm to human health for those who work with waste plastics (end of engineered life, after-use products) and those who are affected by plastic waste processing activities, including the controlled operations of plastics recyclers. On the basis of the evidence summarised, we derived prevalent risk scenarios of hazard-pathway-receptor combinations. 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.

Plastic waste processing activities have resulted in several negative health and safety outcomes. Whereas the majority of these negative outcomes occur in LIMICs, there is, for instance, evidence of small levels of contamination of plastic products available for sale in HICs, by VOCs, BFRs, phthalates, and PTEs, for which the likely source is recycled plastic content that has not undergone stringent source control. For these 'legacy substances', the relative risk scores were generally low, because in most cases, concentrations of most were far below limits imposed by the EU (for instance). Nonetheless, 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. The recycling part of our circular economy does not necessarily provide for safe and final sinks for substances of concern in plastics.

Whereas the potential pathway for legacy substances in plastic is mainly through migration to surface or abrasion under normal use conditions, heating plastics exacerbates the release of some potentially hazardous substances into the environment, and increases the risk of them entering the human body. Extrusion in plastics reprocessing facilities can result in this type of thermal release, and some limited evidence from bodily tissue and several environmental compartments has indicated that workers had been exposed to BFRs, phthalates, VOCs and PAHs in several LIMIC

contexts where safe systems of work were lacking. Very little information to evidence harmful atmospheric emissions from extrusion in HICs was revealed in this research. However, the stringent health and safety regulation and enforcement in these contexts mean that it is more likely than not, that safe systems of work are in operation, alongside engineering controls such as respiratory protective equipment and local exhaust ventilation.

Conversely in some LIMIC contexts, we identified evidence to indicate that no such controls are in place. Several studies showed emission control by just passive ventilation, through open doors and windows followed by dilution and dispersion in the atmosphere. For some LIMIC secondary plastics reprocessors, particularly those that process polymers such as ABS, PVC, PS, PC-ABS and K-resin, either intentionally or as contaminants alongside other plastics, this lack of control could result in the exposure of their workforce to harmful concentrations of VOCs, phthalates and PAHs. Whereas these substances can be easily anticipated according to the polymer being processed, when unknown legacy substances are allowed to enter the secondary plastics value chain, the risks to workers and surrounding population are less predictable. Indirectly, we have revealed evidence that may indicate the release of BFRs into the atmosphere and other surrounding environmental compartments from plastics reprocessing operations and in which case would also expose directly the workers in those establishments.

A circular economy with business as usual control over legacy substances, and operated in loosely controlled and non-traceable globalised supply chains, could result in increased dispersion of substances of concern all over the world. Resultant risks to human health may be comparatively small, yet not sufficiently quantified to be dismissed. Workers can be exposed to harmful emissions from the extrusion of certain recycled plastics if pollution control measures are not in place, which is evidently still common in low and middle income country settings. Such risks are exacerbated of the provenance of the plastics to be recycled cannot be reliably determined, which is often the case, particularly in the Global South.

CRediT author statement

Ed Cook: Conceptualization; Data curation; Formal Analysis; Investigation; Methodology; Project administration; Resources; Validation; Visualization; Writing – original draft; Writing – review & editing. **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. **Michiel Derks:** Formal Analysis; Data curation; Investigation.

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