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# HAZARDOUS PROPERTIES OF PLASTICISERS THAT MAY HINDER THE RECYCLING OF SOFTENED PLASTICS

Pierre Hennebert \*

Ineris (French National Institute for Industrial Environment and Risks), CS 10440, F-13592 Aix-en-Provence Cedex 03, France

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## ABSTRACT

Plasticisers transform rigid polymers, especially PVC, into flexible and useful material, typically at 10-35% concentration. Four phthalate plasticisers are now banned in the EU (maximum concentration in products of 0.1%). Are other plasticisers, used in concentrations that make a product waste, unsafe? The hazardous properties of plasticisers used in the EU (Plastic Additives Initiative list) were collected from the ECHA registration site. Eight plasticisers (=12% of 69) are either skin sensitizers (2 substances) and under evaluation by ECHA (7 substances), with a potential ban at the end of the evaluation for persistence, bioaccumulation and toxicity (PBT), endocrine disruption (ED) and as substances of very high concern (SVHC). Seventeen (=25% of 69) are used at a concentration that makes the plastic hazardous when it becomes waste. The sorting and management options of these additivated plastics are discussed. The recycling of these hazardous wastes is not prohibited. In the short-term recycling phase in modern industrial plants, there is a low emission of these additives, which is controlled by occupational safety and environmental regulations. On the other hand, the long-term low-quality management such as littering (with weathering and fragmentation) and landfilling (with the emission of degradable products in case of phthalates) scatter these substances. The plastics containing "legacy" banned additives must be phased out. But the plastics with compounds at hazardous concentration should be recycled in controlled recycling loop. They should be managed by a risk approach, like the products they were and the new products that they will become.

## 1. INTRODUCTION

Plasticisers transform rigid polymers, especially PVC, into flexible material, enormously increasing the possibilities of applications in construction, automotive, health, cables, floor, toys and packaging (European plasticisers 2022). Their use is generalized. The EU PVC industry has committed to recycle 900 kt of PVC in 2025 and 1 Mt in 2030 (Vinylplus 2022), with 42% of softened PVC. Do these plastics contain "legacy" substances (substances authorized at the time of manufacture but restricted today), like some brominated flame retardants that have been with time classified persistent organic pollutants (POPs) by the Stockholm Convention and banned (detailed review in Sharkey et al. 2020)? Similarly, some low molecular weight phthalate plasticisers like di-ethyl-hexyl phthalate are reassessed with time as hazardous and are substituted by high molecular weight phthalates (Vinylplus 2021). Are some plasticisers used in concentration that makes, according to the waste classification, softened plastic waste hazardous? How to manage these wastes, which were authorized products, during the recycling steps?

To answer to these questions, the chemical classification of all the plasticisers officially used in the EU has been looked for and their functional concentration (the concentration at which they are added) used to calculate if the resulting plastics are hazardous according to the hazardous waste classification. The recycling, sorting and management options of these additivated plastics, especially in modern industrial recycling loops, are then discussed. The raising the question of an approach of hazardous substances by risk management rather than hazard in modern facilities in a circular economy is discussed with more details.

## 2. MATERIAL AND METHODS

### 2.1 The list of plasticisers used in the EU

"An additive was defined according to Article 3(7) of Commission Regulation (EU) No 10/2011 on plastic food contact materials, meaning a substance which is intentionally added to plastics to achieve a physical or chemical effect during processing of the plastic or in the final material or article, and which is intended to be present in the final



material or article. In the context of plastic additives initiative, pigments were also considered under this definition” (ECHA 2021a). That definition is in fact extended to all applications of plastics. The Plastic Additives Initiative (PAI), a collaboration between the European Chemicals Agency - ECHA and the plastics industries delivered in 2019 a list of 418 additives currently used in products in the EU, along with their function(s), the polymer(s) they improve, and their functional concentration(s) (ECHA 2021a). The excel file is no longer available, but the list of additives by function is available (with polymer and functional concentration) and a file can be easily reconstructed from the different screens of ECHA (2021a).

For the 69 plasticisers of the list, the hazardous properties of human toxicity and ecotoxicity were collected from the ECHA open-access registration site of chemicals in the EU (ECHA 2021b). When their functional concentration is mentioned, it is compared with the concentration that makes a waste hazardous for the 15 hazard properties of waste (EU 2014, 2017).

## 2.2 Properties of plasticisers and hazard classification of waste

The self-reported chemical properties of substances on human health and the environment are found in their European Chemicals Agency (ECHA) registration dossier (<https://echa.europa.eu/information-on-chemicals>). The “Brief Profiles” are a practical summary of the dossier. Some hazards are graduated from level 1 (high) to level 4 (low). For some substances, the ECHA mentions its own “harmonised” classification, or indicates that a re-assessment is in progress. The hazard statement that has been

attributed to at least one plasticiser, and the concentration that makes a waste hazardous for a hazard property with these hazard statements are presented in Table 1. The hazard classification of the additivated plastics as waste is done according to the EU regulations (EU 2014, EU 2018) with maximum concentration for some properties and (weighted) summation of concentration for other properties (HP 4, HP 6, HP 8, HP 14). A synthesis of waste classification is presented in Hennebert (2019a). It has been supposed that only one plasticiser is used in a plastic compound. The eventual other additives are not known and hence their properties and concentration have not been taken into account.

### 2.2.1 The case of the concentration limit of substances classified H400 that renders a product and a waste hazardous

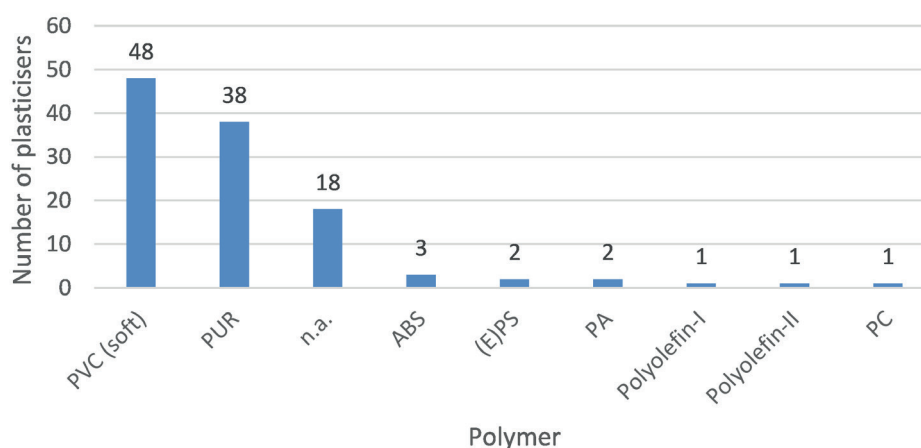
For products (articles and mixtures), for acute and chronic ecotoxicity, M-factors specific to the most ecotoxic substances are multipliers of the concentration of H400 and H410 substances in the weighted sum that calculates if a concentration limit is exceeded. The value of these M-factors rank from 1 to 1 000 000 (this latter case for some pesticides) and are derived from ecotoxicological tests of these substances, as presented in the EU Regulation on classification, labelling and packaging of substances and mixtures, in short CLP (EU 2008). That system avoids multiple concentration limits. In the CLP, the classification formulas of mixtures are: A mixture of substances (a product) is classified aquatic acute or aquatic chronic if:

$$\sum (M_{acute} \times C_{H400}) \geq 25\% \text{ (Ecotoxic Acute Category 1)}$$

**TABLE 1:** The hazard statements of the plasticisers in the ECHA dossiers, the concentration that makes a waste hazardous (by decreasing concentration) and the related waste hazard properties.

Hazard statement	Hazard statement code - HSC	Concentration that makes a waste hazardous	Waste Hazard Property (HP)
Specific Target Organ Toxicity, Single Exposure 3	H335	Not used in waste classification, no concentration limit in product classification	No waste HP
Acute Toxicity 4 (Oral)	H302	25%	HP 6 Acute toxicity
Aquatic chronic 3	H412	25%	HP 14 Ecotoxic
Aquatic chronic 4	H413	25%	HP 14 Ecotoxic
Skin irritant 2	H315	20%	HP 4 Irritant
Eye irritant 2	H319	20%	HP 4 Irritant
Skin sensitizing 1	H317	10%	HP 13 Sensitizing
Eye damage 1	H318	10%	HP 4 Irritant
Specific Target Organ Toxicity, Single Exposure 2	H371	10%	HP 5 Single target organ toxicity
Specific Target Organ Toxicity, Repeated Exposure 2	H373	10%	HP 5 Single target organ toxicity
Acute toxic 3 (Oral)	H301	5%	HP 6 Acute toxicity
Reprotoxic 2	H361	3%	HP 10 Toxic for reproduction
Aquatic chronic 2	H411	2.50%	HP 14 Ecotoxic
Reprotoxic 1A and 1B	H360	0.30%	HP 10 Toxic for reproduction
Aquatic acute 1	H400	Waste classification 25%, Product classification here 0.25%	HP 14 Ecotoxic
Aquatic chronic 1	H410	0.25%	HP 14 Ecotoxic

## Plasticisers by polymer (114 combinations of 69 plasticisers and 9 polymers)



**FIGURE 1:** The use of plasticisers by polymer (calculated from the Plastic Additives Initiative - PAI) [n.a.: not available in the PAI files; ABS: acrylonitrile butadiene styrene; PA: polyamide ; PC: polycarbonate ;Polyolefin-I: polyethylene group; Polyolefin-II: polypropylene; (E)PS: (expanded) polystyrene; PUR: polyurethane; PVC (soft): polyvinylchloride (with plasticisers)].

- $\sum [(M_{\text{chronic}} \times 10 \times C_{\text{H410}}) + (C_{\text{H411}})] \geq 25\%$  (Ecotoxic Chronic Category 2)
- $\sum [(M_{\text{chronic}} \times 100 \times C_{\text{H410}}) + (10 \times C_{\text{H411}}) + (C_{\text{H412}})] \geq 25\%$  (Ecotoxic Chronic Category 3)
- $\sum (C_{\text{H410}} + C_{\text{H411}} + C_{\text{H412}} + C_{\text{H413}}) \geq 25\%$  (Ecotoxic Chronic Category 4)

with  $\sum$  meaning sum and  $C_{\text{H400}}$  = concentration of substances with the hazard statement code H400, etc.

These M-factors are not used in waste classification (EU 2017), leading to an unrealistic concentration limit of 25% for acute ecotoxicity and more realistic 0.25% limit for chronic ecotoxicity. A waste (a mixture of substances) is ecotoxic if:

- $\sum C_{\text{H400}} \geq 25\%$ ; or
- $\sum [(100 \times C_{\text{H410}}) + (10 \times C_{\text{H411}}) + (C_{\text{H412}})] \geq 25\%$ ; or
- $\sum (C_{\text{H410}} + C_{\text{H411}} + C_{\text{H412}} + C_{\text{H413}}) \geq 25\%$ .

Compared to products, the acute ecotoxicity is underestimated by a Macute factor. Compared to products (chronic ecotoxicity level 2), the chronic ecotoxicity of waste is overestimated by a factor of 10 for brominated flame retardants ( $M_{\text{chronic}}$  factors = 1) and underestimated by a factor of 10 or more for instance for plastic packaging in which pesticides ( $M_{\text{chronic}}$  factors = 10, 100 and more) have diffused to concentrations >250 mg/kg (Eras et al. 2017, Jin et al. 2018), making them ecotoxic chronic level 1 (multiplying  $M_{\text{chronic}}$ -factor of concentration = 1 000) or 2 (multiplying  $M_{\text{chronic}}$ -factor = 100) in the product classification, but not in the waste classification. A more complete list of substances can be found in Hennebert (2019).

In line with the principles of the circular economy, discrepancies between products and waste should be avoided. For this reason, for four H400 additives, critical for hazard assessment, the calculation of hazard by the product methods (including the M-factors) are presented in the Results in parallel with the waste method.

## 3. RESULTS AND DISCUSSION

### 3.1 The plasticisers and their combination with polymers

The plasticisers are frequently used as they are in the third rank of substance/polymer combination, after the classes “pigments agents” (979 substance-polymer combinations) and “other functions” (126 combinations) and before the flame retardants class (61 combinations) (Table S1). Sixty-nine (69) plasticisers are used in 9 polymers families, delivering 114 combinations (Table S1, Figure 1). Polyvinylchloride – PVC and polyurethane – PUR are the main softened polymers. It should be noted that four plasticisers can also be flame retardants and that double function is considered in the table. The plasticisers are clearly used at higher concentration than other additives (data in Hennebert 2021): the mean concentration is 21%, the median and maximum concentrations are 35% (with 38 times that maximum concentration out of 44 available concentration data, being a frequency of 86%).

### 3.2 Assessment by the product method of the concentration limit that makes a waste hazardous for two H400 plasticisers

As presented below, zinc bis (dihydrogenophosphate) is classified H400 and triphenyl phosphate is classified H400 and H410 (Table 5, Group 5). To avoid using a concentration limit of 25% for these H400 substances, the product classification of the CLP has been also used. Acute and chronic multiplying M-factors must be assessed. A summary of this stepwise method is available in Hennebert (2019). The hazard statement codes HSC and the M-factors are attributed to substances from experimental laboratory ecotoxicological standardized tests. The ecotoxic concentrations (from the ECHA dossier of these substances) and the resulting M-factors for these two plasticisers are presented at Table S2. The resulting M-factors for acute and chronic ecotoxicity are 1.

As M-factors of these two plasticisers = 1, a mixture containing these plasticisers is ecotoxic acute and chronic level 3 if the concentration of substances with HSC H400 is  $\geq 0.25\%$  and/or if the concentration of substances with HSC H410 is  $\geq 0.25\%$ . To be consistent with products, it is proposed to use as concentration limit for HP 14 'Ecotoxic' a concentration of 0.25% for acute and chronic ecotoxicity (Table 5), in parallel with the concentration limit for waste of 25% for the sum of H400 substances.

### 3.3 Groups of plasticisers by the level of information in the ECHA dossiers

When a "harmonized" (officially classified by expert groups) classification is available, it has been used. For the other substances, there can be up to 1500 notifiers for one substance. Notifiers are marketers (producers or importers into the EU) of the substance. Some plasticisers are not classified (no hazard statement codes are attributed) by some notifiers, meaning that there is no hazard for these notifiers, while other notifiers may have attributed hazard statement(s) and they are used here if they have been declared by more than 10% of the notifiers. If declared by lower than 10%, these HSC have not been taken into account for calculation of the hazard and are bracketed in the Table 4 to Table 6. Some substances are with time under reassessment by ECHA and these re-classification works are mentioned by substance in the tables below. Once reassessment achieved, the new "harmonized" classification must be used.

As presented in detail below, the 69 declared plasticisers can be organized in 6 groups of level of information and classification:

- Group 1 has no CAS number and Group 2 has a CAS number, but both are not notified;
- Group 3 are substances with CAS number and that are notified as non-hazardous (without any hazard statement codes);
- Group 4 are substances that are notified with or without hazard statement codes in the CLP system, depending on the notifiers;
- Group 5 are substances that are notified with hazard statement code(s) in the REACH and CLP system, but for which no "harmonized" classification is available;
- Group 6 are substances that are notified with hazard statement code(s) in the REACH and CLP system, and for which a harmonized classification is available.

All the substances of Groups 2 to 6 have a Brief Profile (a synthetic dossier) in the ECHA database.

#### 3.3.1 Plasticisers that are not notified or notified without hazard statement(s) (Groups 1 to 3)

The groups 1 to 3 are presented in Table 2.

Groups 1 and 2 are groups of chemicals that are not notified in the ECHA database. Their dossiers should be completed.

Group 3 includes 38 non-hazardous substances, including some phthalates. Nevertheless 7 substances of Group 3 have no hazard statement but have regulatory

obligation(s) in different pieces of legislation (mainly regulation on plastic materials and articles intended to come into contact with food), with a controlled total or mobile concentration.

#### 3.3.2 Plasticisers that are notified with hazard statement(s) (Groups 4 to 6)

The groups 4 to 6 are presented in Table 3 and organized by presence of hazard statement in the CLP system (Group 4), in the REACH and CLP system (Group 5) and for which a harmonized classification is available (Group 6).

The two phthalic substances with an asterisk (\*) in Table 5 can be used at a maximum concentration of 0.1% by weight of the plasticised material (Regulation EU 2018/2005). With that concentration, the material is not hazardous, according to the waste classification. Two other substances, benzyl butyl phthalate (BBP) (CAS No.: 85-68-7, EC No.: 201-622-7) and diisobutyl phthalate (DIBP) (CAS No.: 84-69-5, EC No.: 201-553-2) are included in the EU 2018/2005 regulation but are not found in the PAI list of 2019 and are phased out.

For Group 4 (classified with CLP), 4 substances out of 14 are used at minimal concentration that makes the additivated plastic hazardous (according to the waste classification), and 7 substances are used at maximal concentration that makes the plastic hazardous. For Groups 5 (classified with CLP and REACH) and 6 (harmonized classification), 5 and all the 3 substances are used at concentration that makes the additivated plastic hazardous. These results are synthesised in the next section.

For the classification of substances, one can conclude either that a better documented substance is more frequently hazardous, or that the most hazardous substances have been better characterised. It can also be interpreted that the 4 non-notified substances must be urgently notified. For the discussion of the use of concentration limit of waste or of product for ecotoxic acute H400 substances (first subsection of the Results section), zinc bis(dihydrogen phosphate), triphenyl phosphate, [phenol, isopropylated, phosphate (3:1)] and C14-17 chloroalkanes are classified hazardous by HP 14 by the product concentration limit of 0.25%, but also by their HSC H411 (waste limit of 2.5% with functional concentrations of 10-35%) and H410 (waste limit of 0.25% with functional concentration of 2%), respectively. As these substances are also ecotoxic chronic, the use of the product concentration limit for acute ecotoxicity does not change the global classification for HP 14.

### 3.4 Synthesis of classification of plasticisers for hazardous concentrations and for re-assessment by the ECHA

As a synthesis, for the 69 plasticisers (Table 4),

- 4 plasticisers have no dossier or an empty dossier (no notifications by declarant(s))
- 38 plasticisers have a dossier with notification as non-hazardous
- 23 plasticisers have a dossier with notification of the pure (100%) substance as hazardous
- 17 plasticisers (= 25%) are used at concentrations that

**TABLE 2:** The groups of plasticisers with no information, or without notified hazard statement. FC % = functional concentration of the additive in the plastic(s) in % (w/w), OBL = These substances have regulatory obligations in different pieces of legislation.

Plasticiser	CAS no	EC list no	REACH registration	CLP notifications	Harmonised Classification	FC %	OBL
<b>Group 1: No CAS number, not in the ECHA chemicals database</b>							
Reaction mass of benzyl 2-ethylhexyl adipate, bis (2-ethylhexyl) adipate, dibenzyl adipate	No number					10-35	
Reaction mass of: 1-[2-(benzoyloxy)propoxy]propan-2-yl benzoate and 2-[2-(benzoyloxy)ethoxy]ethyl benzoate	No number						
Reaction mass of: 2-[2-(benzoyloxy)ethoxy]ethyl benzoate, 1-[2-(benzoyloxy)propoxy]propan-2-yl benzoate and 2-[2-(benzoyloxy)ethoxy]ethyl benzoate	No number						
<b>Group 2: No notification in ECHA database</b>							
C14-17 alkanes, sec-mono- and disulfonic acids, phenyl esters	No number	701-257-8	Not notified	Not notified	Not notified		
<b>Group 3: Notified, not classified in REACH and CLP, neither in harmonised classification</b>							
1,2,4-Benzenetricarboxylic acid, mixed decyl and octyl triesters	90218-76-1	290-754-9	NC	NC			
1,2,4-Benzenetricarboxylic acid, tri-C9-11-alkyl esters	94279-36-4	304-780-6	NC	NC		10-35	
1,2-Benzenedicarboxylic acid, di-C11-14-branched alkyl esters, C13-rich	68515-47-9	271-089-3	NC	NC		10-35	
1,2-Benzenedicarboxylic acid, di-C16-18-alkyl esters	90193-76-3	290-580-3	NC	NC		10-35	
1,3-Propanediol, 2,2-dimethyl-, 1,3-dibenzoate	4196-89-8	224-081-9	NC	NC		10-35	
2,2-bis[(1-oxopentyl)oxymethyl]propane-1,3-diyl divalerate	15834-04-5	239-937-7	NC	NC		10-35	
2,2'-ethylenedioxydiethyl bis(2-ethylhexanoate)	94-28-0	202-319-2	NC	NC			
Amides, C16-C18 (even), N,N'-ethylenebis	68390-94-3	931-299-4	NC	NC		1	
bis(2-(2-butoxyethoxy)ethyl) adipate	141-17-3	205-465-5	NC	NC			
Bis(2-propylheptyl) phthalate	53306-54-0	258-469-4	NC	NC		10-35	
bis(decyl and/or dodecyl) benzene-1,2-dicarboxylate	No number	931-251-2	NC	NC		10-35	
Decanedioic acid, 1,10-diisodecyl ester, Diisodecyl sebacate	28473-19-0	249-047-0	NC	NC			
Dihexyl adipate	110-33-8	203-757-7	NC	NC		10-35	
Diisodecyl azelate	28472-97-1	249-044-4	NC	NC			
Diisononyladipate	33703-08-1	251-646-7	NC	NC		10-35	
Diisotridecyl adipate	26401-35-4	247-660-8	NC	NC		10-35	
Diisotridecyl phthalate	27253-26-5	248-368-3	NC	NC		10-35	
Diundecyl phthalate, branched and linear	85507-79-5	287-401-6	NC	NC			
Dodecanoic acid, ester with 1,2,3-propanetriol, acetylated	91744-35-3	294-597-7	NC	NC		10-35	
Esterification products of 1,3-dioxo-2-benzofuran-5-carboxylic acid with nonan-1-ol	1689576-55-3	941-303-6	NC	NC			
Fatty acids, C16-18, C12-18-alkyl esters	95912-87-1	306-083-2	NC	NC		0.5	
Fatty acids, C18-unsatd., dimers, 2-ethylhexyl esters	68334-05-4	500-204-4	NC	NC			
Fatty acids, C8-10, C12-18-alkyl esters	95912-86-0	306-082-7	NC	NC			
Glycerides, C12-18	67701-26-2	266-944-2	NC	NC			
Glycerides, C14-18	67701-27-3	266-945-8	NC	NC			
Isobutyric acid, monoester with 2,2,4-trimethylpentane-1,3-diol	25265-77-4	246-771-9	NC	NC			
Isononanoic acid, C16-18 (even numbered)-alkyl esters	111937-03-2	601-141-6	NC	NC			
Isosorbide Diesters; Fatty acids, C8-10, diesters with 1,4:3,6-dianhydro-D-glucitol	1215036-04-6	700-073-5	NC	NC			
Linseed oil, epoxidized	8016-11-3	232-401-3	NC	NC		10-35	
Oxydiethylene dibenzoate	120-55-8	204-407-6	NC	NC			
Trioctyl benzene-1,2,4-tricarboxylate	89-04-3	201-877-4	NC	NC			
1,2-Cyclohexanedicarboxylic acid, diisononyl ester, reaction products of hydrogenation of di-isononylphthalates (n-butenes based); Di-isononyl cyclohexanoate	166412-78-8	431-890-2	NC	NC		10-35	Yes
Bis(2-ethylhexyl) terephthalate	6422-86-2	229-176-9	NC	NC			Yes
Diisononylphthalate	28553-12-0; 68515-48-0	249-079-5	NC	NC		10-35	Yes
Soybean oil, epoxidized	8013-07-8	232-391-0	NC	NC			Yes
Triacetin	102-76-1	203-051-9	NC	NC		10	Yes
Tributyl-O-Acetyl citrate	77-90-7	201-067-0	NC	NC		10-35	Yes
Triethyl citrate	77-93-0	201-070-7	NC	NC		10-35	Yes

Notes for the table: NC = not classified (no hazard statement codes attributed by the notifiers); FC % = functional concentration of the additive in the plastic(s) in % (w/w)  
OBL = These substances have regulatory obligations in different pieces of legislation.

make the plastics hazardous (according to the waste classification). These 17 plasticisers are listed in Table 5 (second to last column colored in pink)

- Of which 8 plasticisers (= 12%) are either skin sensitizing (2 substances), and under assessment by ECHA, with a potential ban at the end of the evaluation for PBT, ED and SVHC issues (7 substances). These 8 substances are listed in Table 3 (column "harmonized classification").

### 3.5 Recycling of hazardous plasticized polymers

The 17 hazardous plasticisers used at concentrations that make the plastics hazardous are added mainly in PVC (9 substances with 4 under assessment, of a total of 44 plasticisers) and in PU (9 substances with 3 under assessment, of a total of 34 plasticisers) (Table S3), and the recycling concerns will focus on these polymers. They are not used in Polyolefin-I, Polyolefin-II and PC.

The recycling of PVC (hard and softened) has reached 731 kt in 2020 in the EU, and the industry has committed to recycle 900 kt in 2025 and 1 Mt in 2030 (Vinylplus 2022). The PVC comes from: window profiles and related products (48%), flexible PVC and films (including roofing and waterproofing membranes, flooring, coated fabrics, flexible and rigid films) (26%), electrical cables (16%), pipes and fittings (10%). The total of softened PVC is not known but could be about 42%. The recycled PVC is used in windows and profiles (35%), traffic management (15%), pipes (13%), floor covering (10%), thermoformed sheets (5%), roof covering (4%), others (4%), horticultural and stable equipment (3%), another cycle (3%), compounding (3%), export (3%), coils and mandrels (2%) (same source). PVC is not reused in food-contact articles. The recycling of these plastics must be controlled, to avoid for example plasticisers in food packaging materials (e.g. Vapenka et al., 2016). As for all material, the preferred option for PVC management is separate collection, feasible in construction and demolition waste, electrical cables, pipes. Softened PVC has a density between 1.15 and 1.35 kg/L. In simple density separation systems, it will not be separated of hard PVC (1.30 – 1.45 kg/L) and brominated flame retarded plastics BFR HIPS (1.15 – 1.18 kg/L) and BFR ABS (1.15 – 1.22 kg/L), nor of PMMA, PC and PET (Haarman et al. 2020), and could be lost. Additional steps of separation with other physical or optical principles are necessary.

The sorting of plastics faces many challenges: plastic waste streams can be mono or mixed plastic, clean or contaminated, multi-layered, multi-material or composite plastic or whether containing legacy additives (ECHA 2021). For instance, recycling composite materials can be challenging due to their inherently heterogeneous nature. To our knowledge, there is no on-line optical method available for sorting softened and non-softened PVC or PU. The plastics containing legacy additives must face a conflict of objectives, namely saving polymeric resources versus phase-out of hazardous compounds (Wagner and Schlummer 2020). With near infrared (NIR) light, non-black PVC can be separated from the other polymers. Today there is no online sorting method to separate banned low molecular weight phthalates from the other phthalates and the other

plasticisers. The only solution nowadays is selective demolition and manual sorting of homogeneous batches of used articles before they join the waste stream.

The sorted PVC is recycled mechanically, and research is active on chemical recycling. Numerous research and development projects are ongoing (overview in Vinylplus 2022), including recovery of HCl and recovery of chlorine after incineration. An objective could be the selective extraction of legacy additives: "...the first promising plasticizer batch-extraction tests were performed at the Fraunhofer Institute for Chemical Technology ([www.ict.fraunhofer.de](http://www.ict.fraunhofer.de)) on PVC dry blends and PVC sheets (mostly containing mainly DEHP). The extraction achieved good yields of over 70%. » (Vinylplus 2022). Another promising initiative is the EU funded Circular Flooring consortium: "Circular Flooring aims to fully recycle waste PVC flooring by applying the CreaSolv® Process for PVC recovery with simultaneous removal of undesired legacy phthalates which are subjected to catalytic hydrogenation, finally yielding revalorised safe plasticisers." (Circular Flooring 2022).

Substitution of low molecular weight classified orthophthalates by higher molecular weight orthophthalates, cyclohexanoates, terephthalates and other plasticisers in Europe is nearly 100% in 2020 (<https://www.europeanplasticisers.eu/>).

### 3.6 Management of hazardous plasticised polymers by risk during collection and preparation for reuse in modern facilities

To produce plasticised plastic articles, plasticisers are synthesized in the chemical industry. They are transported to formulators, which combine polymers with different additives. Granulated compounds are transported to article manufacturers. After production, the items are transported and used as building materials or consumer items. The emission of plasticiser to humans and/or the environment can occur during the synthesis, transport and formulation of granules by dust emission. The plasticisers are mixed into a polymer matrix and have been designed to not diffuse to the surface or not evaporate. There are no or very few emissions during the life of the item with normal use. These emissions are controlled by the product's regulations. Exposure of workers is controlled by occupational safety and health regulations. The emission of granules containing plasticisers can occur by accidental spillage.

Wasted products are collected, transported, sorted, prepared for reuse, recomposed, regranulated and delivered to plastic processors for manufacture of new products from recycled materials. In modern installations of recycling companies, there are few emissions for humans or the environment. Any dust emissions during grinding are controlled. There are phthalates in plastic recyclates up to some g/kg (for instance Pivnenko et al. 2016).

Non-reactive additives interact with the polymers via weak non-covalent bonds (ECHA 2021c). As a result, they can leach into the environment during the intended life cycle and the waste phase of the plastic product (Wagner and Schlummer, 2020). In case of littering or landfilling of waste, the alteration of the polymer release in the long

**TABLE 3:** The groups of plasticisers with notified hazard statement(s) and harmonized classification, by decreasing concentration making the waste hazardous (pink colored cells). Colors of Hazard Properties is conventional and for global overview only: NH 'Non-hazardous' = green, HP 4 'Irritant' (low intensity)= black, HP 5 'Specific toxicity', HP 6 'Toxic', HP 10 'Toxic for reproduction' = red, HP 14 'Ecotoxic' = blue. FC % = functional concentration of the additive in the plastic(s) in % (w/w), OBL = These substances have regulatory obligations in different pieces of legislation.

Plasticisers / Groups	CAS no	CLP notifications	Harmonised Classification	Lowest Hazardous concentration	FC %	Hazardous by min FC	Hazardous by max FC	OBL
Group 4: Classified with CLP								
Paraffin waxes and Hydrocarbon waxes, chloro (also flame retardant)	63449-39-8	NC, (H319), (H400), (H362)						
Decanedioic acid, 1,10-bis (2-ethylhexyl) ester	122-62-3	NC, (H302)			10-35	NH	NH	
Reaction mass of tris(2-chloropropyl) phosphate and tris(2-chloro-1-methylethyl) phosphate and Phosphoric acid, bis(2-chloro-1-methylethyl) 2-chloropropyl ester and Phosphoric acid, 2-chloro-1-methylethyl bis(2-chloropropyl) ester (also flame retardant)	No number, EC 911-815-4	H302			15	NH	NH	
Bis(2-ethylhexyl) adipate	103-23-1	NC, (H400), (H410)			10-35	NH	NH	Yes
Di-C8-10-Branched alkyl esters, C9-rich	68515-48-0	NC, (H400)			10-35	NH	NH	Yes
Dimethyl sebacate	106-79-6	NC, (H318), (H412)			10-35	NH	NH	
Di-C9-11-Branched alkyl esters, C10-rich; Di-isodecyl phthalate	68515-49-1	NC, (H319), (H315)			10-35	NH	NH	Yes
Isodecyl diphenyl phosphate	29761-21-5	H413, NC, (H400), (H410)		25%	10-35	NH	NH	
Bis(tridecyl) adipate	16958-92-2	NC, (H317)	Ss		10-35	NH	NH	
Glycerides, C8-18 and C18-unsatd. mono- and di-, acetates	91052-13-0	NC, H412		25%	10-35	NH	HP 14	
Di-n-butyl terephthalate	1962-75-0	NC, H315, H319, H412		20%; 25%	10-35	NH	HP 4, HP 14	
Dibutyl sebacate	109-43-3	NC, H319, H315, H335		20%	10-35	NH	HP 4	
Fatty acids, C8-18 and C18-unsatd., esters with trimethylolpropane	85186-89-6	NC, H315, H318		10%	10-35	HP 4	HP 4	
Tributyl citrate	77-94-1	NC, H318, (H400)		10%	10-35	HP 4	HP 4	
Tris(2-ethylhexyl) benzene-1,2,4-tricarboxylate	3319-31-1	NC, H361, (H319), (H315), (H335), (H413)	PBT, ED	3%	35	HP 10	HP 10	Yes
Dibutyl adipate	105-99-7	NC, H411		2.50%	10-35	HP 14	HP 14	Yes
Group 5: Classified in REACH and CLP								
1,2,3-trihexyl 2-(butanoyloxy)propane-1,2,3-tricarboxylate	82469-79-2	NC, (H400), (H411)			10-35	NH	NH	
N-butylbenzenesulphonamide; N-butylbenzenesulfonamide	3622-84-2	H412, H373, NC, (H371), (H302)		10%	10-35	HP 5	HP 5	
Nonylbenzoate, branched and linear	670241-72-2	H411, H361		2.50%	10-35	HP 14, HP 10	HP 14, HP 10	
Oxydipropyl dibenzoate	27138-31-4	H412, NC, H411, (H319), (H361)		2.50%	10-35	HP 14	HP 14	
Zinc bis(dihydrogen phosphate)	13598-37-3	H400, H302, H411, NC		0.25%; 2.5%	10-35	HP 14	HP 14, HP 6	
3 Phenol, isopropylated, phosphate (3:1) (also flame retardant)	68937-41-7	H361, H373, H317, H411, H410, H413	PBT assessment	0.25%	15-35	HP 14, HP 10, HP 5	HP 14, HP 10, HP 5	
Triphenyl phosphate	115-86-6	H400, H410, H411, (H413)	ED	0.25%; 0.25%	2	HP 14	HP 14	
Group 6: (Classified in REACH, CLP), Harmonised classification								
2 Alkanes, C14-17, chloro (also flame retardant)	85535-85-9	NC	H400 H410 H362 PBT	0.25%	15	HP 14	HP 14	Yes
Di-2-ethylhexyl phthalate (DEHP)*	117-81-7		H360FD, SVHC, ED	0.30%	2*-35*	HP 10	HP 10	Yes
Dibutyl phthalate (DBP)*	84-74-2		H400, H360Df, SVHC, PBT	0.25% / 0.3%	10*-35*	HP 14, HP 10	HP 14, HP 10	Yes
Di-allyl phthalate	131-17-9		H400, H410, H302, Ss	0.25% / 0.25%	10-35	HP 14	HP 14, HP 6	Yes

Notes for the table: \* = see text; NC = not classified (no hazard statement codes are attributed) by some notifiers; Bracketed hazard statement code HSC = declared by less than 10% of the notifiers, not taken into account for hazard classification; NH = nonhazardous; HP x(x)= hazardous by the hazard property x(x) (details in Table 1); H360FD = Reprotoxic level 1 FD: F May damage fertility, D May damage the unborn child; H360Df = Reprotoxic level 1 Df: D May damage the unborn child, f Suspected of damaging fertility; Ss = skin sensitizing; SVHC = substance of very high concern; ED = endocrine disruptor; PBT = persistent, bioaccumulable, toxic (potentially future persistent organic pollutant – POP)



term the additives that can be emitted into underground soil and via landfill leachates to water bodies. However, leaching only through molecular diffusion is considered a very slow process, and most substances are released to the environment due to wear-and-tear and pulverisation (Sun et al., 2016). A review on the emission of xenobiotics in landfill leachate has indicated the presence of phthalates (Slack et al. 2005). In another study, leachates from 17 different landfills in Europe were analysed with respect to phthalates and their degradation products (Jonsson et al. 2003). Phthalates were present in the majority of the leachates investigated (monoesters appeared from 1 to 20 µg/L, phthalic acid 2–880 µg/L, parental diesters were observed from 1 to 460 µg/L). All diesters studied are degraded by microorganisms under the landfill conditions. They were released from formulations in a variety of products, including polyvinyl chloride (PVC) species. So, on the long-term in landfill conditions, the phthalates are mobile but also biodegradable. Landfill leachates are in some cases treated in dedicated facilities or municipal treatment plants before being discharged into natural water bodies. Otherwise, specific coagulation and flocculation process, or even Fenton process, are recommended (Zheng et al. 2009, He et al. 2002).

The management of hazardous waste by a risk approach during preparation of secondary raw material in modern installations is risen for many wastes. Many authors conceptualise a legal framework for plastics in circular economy (Van Bruggen et al. 2022, Bening et al. 2021, King and Locock 2022, Paletta et al. 2019, Aurisano et al. 2021, Shamsuyeva and Endres 2021). In the circular economy, hazardous waste, old products, and future products, should be managed by a risk approach. Hazardous products circulate with certain substance restrictions, for a defined use, with information (such as safety data sheet)

and labeling for the user. During the normal intended use, there are controlled emission(s)/exposure(s) of substances hazardous to humans or the environment, and therefore the risk is kept acceptable, as the risk results from the exposure of a target to hazard. During the recycling and use phases, there is no conceptual objection to managing recycled material, possibly containing hazardous substances, differently from virgin material. But in case of abandonment or long-term storage in the environment of an abandoned product then designated as waste, as there is no longer any control of the emission and exposure of the material, the material must be managed by its hazardous properties (Hennebert 2022). The interface between chemicals and waste legislation is a major problem for the envisaged circular economy (Friege et al. 2021). Appropriate risk management tools to control any risks that might arise from the re-using and recycling of hazardous materials should be built (Bodar et al. 2018). These authors recommend connecting the separate legal framework of products and of waste by interlinking with the REACH regulation, with a management by the risk.

### 3.7 Ranking hazard properties of hazardous softened plastics

Regarding the prevalence of hazard properties at the functional concentration of plasticisers (Table 4 and Table 5), plastics containing additives are classified 11 times as hazardous by HP 14, 5 times by HP 10, 4 times by HP 4, 2 times by HP 6, and 2 times per HP 5.

The most frequent hazard property is HP 14 'Ecotoxic'. The ecotoxicity of substances is assessed by adding dissolved or suspended substances to the organism culture medium for aquatic tests, or by adding powder to the organism culture medium for solid or terrestrial tests. The ecotoxicity of a waste is evaluated by calculation (as in

**TABLE 4:** The classification of plasticizers by level of information and the number of plasticizers that make and do not make plastics hazardous.

Group	Classification of plasticisers	Number	Number of Functional Concentration	Plastics Hazardous with documented minimum functional concentration	Plastics Non-Hazardous with documented maximum functional concentration	On-going assessment by ECHA
1	No CAS number, not in the ECHA chemicals database	3	1		If no HSC, n = 3	
2	Entry in ECHA database without notification	1	0		If no HSC, n = 1	
3	Notified, not classified in REACH and CLP, neither in harmonised classification: no HSC	38	20		38	
4	Classified with CLP: HSC	16	15	7	8	1 Ss, 1 PBT ED
5	Classified in REACH and CLP: HSC	7	7	6	1	1 ED, 1 PBT assessment
6	Classified in REACH, CLP and in harmonised classification: HSC	4	4	4*	0	1 Ss, 1 ED, 2 SVHC, 2 PBT
Total		69	47	17 = 25% of 69	51 = 74% of 69	8 = 12% of 69

HSC = hazard statement code; \* DEHP and DBP can be used at a maximum concentration of 0.1% by weight of the plasticised material (Regulation EU 2018/2005) and with that concentration, the material is not hazardous according to the waste classification.

this article) or by aquatic test carried out on different dilutions of leachate (demineralised water extract) and by land test carried out with different additions of solid waste in the culture media of the organisms (Hennebert 2017). For plasticisers, during waste management operations, prohibited litter is the only significant emission pathway to the environment and is not expected to occur in industrial facilities.

The health properties (HP 10 'Toxic to reproduction', HP 6 'Acute toxicity' and HP 5 'Single target organ toxicity') are, at least in the short term, controlled by the absence of exposure during normal use of articles and normal management of end-of-life articles. The property HP 4 'Irritant' (skin irritation and eye damage) is relevant when there is direct exposure with the skin or the eyes in case of powder or dust of pure plasticiser (when handling for manufacturing), but not when the additive is mixed and in molded plastics, and when articles or reprocessed articles are used.

## 4. CONCLUSIONS

With the present data in the ECHA dossiers, 17 plasticisers out of 69 (= 25%) declared in the Plastic Additives Initiative are used in concentration that makes the plastic hazardous when it becomes a waste. Two of them (di-2-ethylhexyl phthalate - DEHP and dibutyl phthalate - DBP) are restricted to <0.1%, which is not a functional concentration. Two other phthalates are restricted but are not found in the Plastic Additives Initiative list of 2019. Of these 17, 8 plasticisers (= 12%) are either skin sensitizing (2 substances), and under assessment by ECHA, with a potential ban at the end of the evaluation for PBT, ED and SVHC issues (7 substances).

These softeners are used mainly in PVC and polyurethane. The recycling of hazardous waste is not prohibited: the plastic with a plasticizer (minus the two restricted) at hazardous concentration can be recycled. In normal recycling conditions and reuse, there is low emission of these additives. These emissions are controlled by occupational safety and products regulations. On the contrary, low quality waste management like litter (with weathering and fragmentation-pulverisation) and landfilling (with long-term emission, but of degradable products in case of phthalates) are emitting these substances.

The plastics containing "legacy" banned additives must be phased out. But the plastics with hazardous compounds at hazardous concentration should be recycled in controlled recycling loop. This means that they should be managed by a risk approach, like the products they were and the products that they will become.

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