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Non-persistent chemicals in polymer and non-polymer products can cause persistent environmental contamination: evidence with DEHP in Europe

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Abstract

Bis(2-ethylhexyl)phthalate (DEHP) is a plasticizer that has been massively used since the second part of the twentieth century by the plastic industry to provide softness properties to PVC. This chemical is considered as toxic to reproduction endocrine disrupting, and a wide range of uses are now forbidden by the EU. Despite these regulations, DEHP is still found to be a widespread contaminant in watersheds in the EU. In this study, we calculate retrospective and prospective scenarios of past and future emissions of DEHP in the environment (water, soil, air) in the EU 28, taking into account the entire lifecycle of the substance, from its production and its inclusion in polymer (mainly PVC) and non-polymer products (adhesive and sealant, ceramic and printing ink) to the recycling and end of life of these products. We develop a stock and flow model based on dynamically estimating the stocks of DEHP present in products on the market. Our results show that the introduction of recent regulations to limit the use of DEHP (that bring a 70% reduction of DEHP contained in products placed on the market in 2020 and 75% in 2040) will not reduce significantly future emissions. This persistence of emissions is explained by the high stocks built in the economy and the long-term presence of soft PVC waste in landfills. Our results suggest that DEHP will remain a cause of environmental contamination many decades after uses have declined and even ceased, and it appears to be too late for market regulation at the market stage to offset the effect of past stock buildup and landfilling.

It is likely that several chemicals that are not considered as persistent and therefore not the focus of international regulations could exhibit the same characteristics. Regulations should avoid possible use patterns that make hazardous chemicals persistent in products, because they have the potential to create long-term and almost irreversible environmental pollution and impacts.

Keywords: Emissions through water, Soil and air, DEHP, Phthalate, Plasticizer, PVC, Regulation, Persistence, Pollution

1 INTRODUCTION

Bis(2-ethylhexyl) phthalate (DEHP) is a plasticizer providing softness to plastic material (almost exclusively to “soft” polyvinyl chloride). DEHP has been used since 1939 in the USA (IARC 2013), and since then, it has been increasingly used worldwide during the second part of the twentieth century. In 1960 DEHP represented 80% of phthalates used globally. Since 2000, the DEHP market is experiencing a downward trend. A first explanation for this trend is its classification as possibly carcinogenic for humans (category 2B) by (IARC 2013). Moreover, the inscription of DEHP as a substance of very high concern (SVHC) and inclusion in Annexes XIV (authorization) and XVII (restriction) under REACH and its recognition as an endocrine disruptor for human health by the EU led to the development by the plastic sector of alternative plasticizers (ECHA 2008). Approximately, 97% of DEHP is used in the production of PVC (Norwegian Institute for Air Research 2007) and is added to PVC in a proportion up to 30%. It can be found in the following PVC products: flooring and wall coverings, coated fabrics, cables, hoses and profiles, moulded products, medical devices, films and sheets, roofing products, car under-coating (wear, rust and corrosion protection), car frames, etc. The remaining (3%) is used in the production of adhesives and sealants, lacquers and paints, printing inks, ceramics, mastics for indoor and outdoor use and other products. Because of its leaching from these products, DEHP has been found repeatedly in the environment. For instance, it has been found to be the most prevalent of phthalate esters in watercourses of the Paris area (Teil M.J. et al. 2007) and to be a widespread contaminant of the Seine watershed soils (Gasperi J. et al. 2018) and of some fish populations (Bonnard M. et al. 2020). The stock of DEHP in products could act as a source of long-term release to the environment, as has been already shown for other chemicals like the persistent, bioaccumulative, and toxic (PBT) decabromodiphenyl ether (Earnshaw M.R. et al. 2013, Brignon J.M. et al. 2018) or for engineered nanomaterials (Song R. et al. 2017).

The objective of this work was to show that this issue also exists for non-PBT chemicals, using the case of DEHP. We therefore estimate retrospective and prospective emissions of DEHP over a large period (1960-2040) in the current limits of the EU 28, based on its uses and its stocks that built in the economy over time. The full lifecycle of DEHP has been considered, from its production to the end of life of relevant products. Another objective was to study the impact on emissions of recent regulations limiting the use of DEHP, such as the restrictions on the use of DEHP adopted in 2011 under the EU REACH Regulation. Adopted restriction under REACH (European Commission 2018) demands that products with a concentration of DEHP equal to or greater than 0.1% by weight of the plasticised material in the product DEHP shall not be placed on the market after 7 July 2020¹. This regulation applies for DEHP individually or in any combination with the following phthalates: dibutyl phthalate (DBP), benzyl phthalate, butyl phthalate (BBP) and diisobutyl phthalate (DIBP). Recycling PVC could have the unintended detrimental effect of prolonging the presence on the market and therefore of DEHP emissions to the environment (Brignon J.M. 2021). We therefore also study the impact on emissions of the impossibility under REACH to recycle PVC without previously removing DEHP.

2 MATERIALS AND METHODS

2.1 DEHP STOCK AND FLOW MODELLING

We develop a stock and flow model based on dynamically estimating the stocks of DEHP present in products on the market and then computing the emissions resulting from this stock. Estimation of products containing DEHP used has been based on data on quantities of products manufactured, imported and exported. Main sources of information for these calculations are regulatory documents, economic studies and scientific papers dealing with emissions of chemicals (European-Chemicals-

¹ This restriction does not apply to products for agricultural or industrial use, medical devices, aircraft, measuring devices for laboratory use, products placed on the market before 7 July 2020 and motor vehicles within the scope of Directive 2007/46/EC placed on the market before 7 January 2024.

Bureau 2008, Lindeboom R.et al. 2009, COWI 2013, Purser D. 2015). Emissions are separately estimated for each environmental compartment: air, soil and water.

We estimate EU-wide DEHP stocks and emissions in the environment for 14 types of products present on the market that contains DEHP, for each stage of its life cycle, starting with the production of DEHP, the use of products containing DEHP, with finally the end of life of products (eventually followed by recycling). Some of the dates of market introduction and lifetimes presented in Table 1 for the 14 types of products considered are assumptions.

Table 1 List of products containing DEHP and main assumptions for stock calculation

Product or material	Examples of products	Type of use	Year of introduction in the market	Lifetime (years)	Consumption trend between 1960 and 2020	Consumption forecast between 2020 and 2040 in function of the regulation scenario
PVC products						
Flooring	Vinyl flooring	Indoor	1975 (Kleijn R.et al. 2000)	15 (Kleijn R.et al. 2000)	Around 70,000 tons per year placed on the market between 1990 and 2002. Strong decrease between 2002 (53,000 tons/year) and 2020 (28,000 tons/year)	Baseline scenario : around 20,000 tons placed on the market per year Regulation scenario : no change with baseline scenario
Wall covering	Vinyl wall coverings	Indoor	1975 (Kleijn R.et al. 2000)	15 (Assumption)	Around 100,000 tons per year placed on the market between 1990 and 2002	Baseline scenario : less than 10,000 tons placed on the market per year Regulation scenario: no wall covering products containing DEHP placed in the market during this period
Coated fabric	Table cloth, clothes Artificial leather , air mattresses, shower curtains, gloves, tarpaulins, curtains, table linen	Indoor and outdoor	1960 (Wikipédia)	10 (Lindeboom R.et al. 2009)	80,000 tons per year placed on the market between 1990 and 2000. Strong decrease between 2000 (64,000 tons/year) and 2020 (11,000 tons/year)	Baseline scenario: around 20,000 tons placed on the market per year Regulation scenario: around 10,000 tons or less placed on the market per year
Film/sheet and coated product	Packaging, tape and self-adhesive foils	Indoor and outdoor	1960 (COWI 2013)	5 (assumption)	Around 70,000 tons placed on the market between 1970 and 2000. Strong decrease between 2000 (68 000 tons/year) and 2020 (19,000 tons/year)	Baseline scenario: around 20,000 tons or less placed on the market per year Regulation scenario: no products placed in the market during this period

Wires and cables	Computer cable, electric cable	Indoor and outdoor	1965 (Kleijn R.et al. 2000)	30 (Kleijn R.et al. 2000)	Around 80,000 tons placed on the market between 1970 and 2000. Very strong decrease between 2000 (76,000 tons/year) and 2018 (18,000 tons/year)	Regulation scenario: no products placed in the market during this period
						Regulation scenario: no products placed in the market during this period
Moulded product	Toys, shoes soles, bathing equipment	Indoor and outdoor	1960 (COWI 2013)	5 (Lindeboom R.et al. 2009)	Around 50,000 tons placed on the market between 1970 and 1990. In 2000 42,000 tons/year has been placed on the market. Small decrease after 2000, in 2020 38,000 tons/year has been placed on the market	Baseline scenario: around 45,000 tons placed on the market between 2020 and 2040
						Regulation scenario: no products placed in the market during this period
Roofing calendered and coil-coated	Calendered roofing material, coil coated roofing material	Outdoor	1960 (COWI 2013)	20 (Lindeboom R.et al. 2009)	Around 5000 tons placed on the market between 1970 and 2000. Decrease between 2000 (5000 tons/years) and 2020 (3000 tons/years)	Baseline scenario: around 5000 tons placed on the market between 2020 and 2040
						Regulation scenario: no change with baseline scenario
Hoses and profiles	Profile of windows, in industry, garden hoses and tubes, moulded products	Indoor and outdoor	1960 (Kleijn R.et al. 2000)	30 (Lindeboom R.et al. 2009)	Around 60,000 tons placed on the market between 1970 and 1990. Very strong decrease between 2000 and 2020. Less than 10,000 tons placed on the market in 2020	Baseline scenario: Less than one ton placed on the market between 2020 and 2040
						Regulation scenario: no change with baseline scenario
Car undercoating	Car undercoating (wear, rust and corrosion protection) for car frame	Outdoor	1960 (COWI 2013)	12 (Lindeboom R.et al. 2009)	Around 7000 tons placed on the market between 1970 and 2000. Decrease between (6000 tons/years) and 2020 (3000 tons/years)	Baseline scenario: around 3000 tons placed on the market between 2020 and 2040
						Regulation scenario: no change with baseline scenario
Adhesive and sealant		Indoor and outdoor	1960 (COWI 2013)	20 (Lindeboom R.et al. 2009)	Around 10,000 tons placed on the market between 1970 and 2000. Decrease between 2000 (10,000 tons/years) and 2020 (6500 tons/years).	Baseline scenario: decrease between 2020 and 2040. Around 5000 tons placed on the market

						Regulation scenario: no change with baseline scenario
Lacquers and paints		Indoor and outdoor	1960 (COWI 2013)	7 (Lindeboom R.et al. 2009)	Around 1500 tons placed on the market between 1970 and 2 000. Decrease between 2000 (1500 tons/years) and 2020 (700 tons/years)	Baseline scenario: decrease between 2020 and 2040. Less than 1 tons placed on the market
						Regulation scenario: no change with baseline scenario
printing ink		Indoor	1960 (COWI 2013)	1 (Lindeboom R.et al. 2009)	Around 1500 tons placed on the market between 1970 and 2 000. Decrease between 2000 (1600 tons/year) and 2020 (800 tons/years)	Baseline scenario: Decrease between 2020 and 2040. Less than 1 tons placed on the market
						Regulation scenario: no change with baseline scenario
Ceramic products		Indoor	1960 (COWI 2013)	20 (Lindeboom R.et al. 2009)	Around 30 tons placed on the market between 1970 and 2000. Small decrease between 2000 (28 tons/years) and 2020 (17 tons/years)	Baseline scenario: decrease between 2020 and 2040. Less than 10 tons placed on the market
						Regulation scenario: no change with baseline scenario

After 2020 two different evolutions of stocks of products are estimated in order to study the impact of a regulation on DEHP uses. All quantities of DEHP are related to the geographical zone corresponding to the EU 28 as in 2020, and as we study DEHP stocks and emissions from 1960 to 2040, the countries involved in the study are not a member of the EU at all times (Fig. 1).

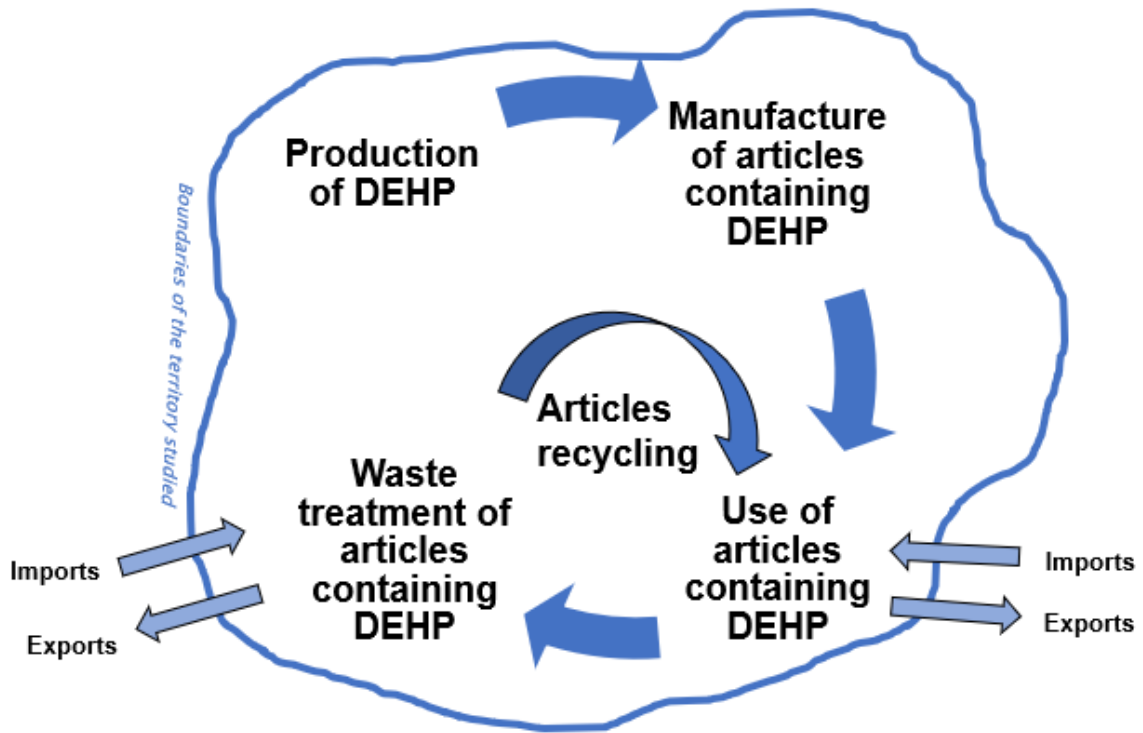


Fig. 1 Life cycle of DEHP and related products

For a given product, emissions are estimated using the general Eq. 1, adapted from (Song R. et al., 2017) and applied to each type of the 14 products considered in this study. Emissions are expressed in tons of DEHP.

$$Emissions (E)[tons] = Emission Factor (EF)[\%] * Stock of DEHP (S)[tons] \quad (1)$$

Emission factors used in Eq. 1 are presented in Table 2 hereafter and all the values used are presented in detail in supplementary material: Suppl mat – Table 4 (EF for the production of DEHP), Suppl mat – Table 5 (EF for the manufacture of products containing DEHP), Suppl mat – Table 6 (EF for the use of products containing DEHP), Suppl mat – Table 7 (EF for the dismantling of products outdoor used) and Suppl mat – Table 8 (EF for end-of-life treatments of products containing DEHP). EF depends on the type of product, its use pattern (indoors vs. outdoors) and on the product life cycle step. To apply this equation, DEHP stocks “S” produced, transformed, used and processed at their end of life (landfilled, incinerated or recycled) in Europe have been estimated between 1945 and 2040 for each type of product with the following equations (2, 3 and 4). Equation 2 estimates additional quantities of DEHP placed on the market through new or recycled products of type “x” during a given year numbered N. Equation 3 is used to quantify the remaining stock of DEHP contained in products of type x, at a year $n > N$, for products that were placed on the market at year N, considering emission from use, and from dislocation only for items with outdoor uses. Finally, Eq. 4 expresses the total stock of DEHP present on the market at year n for product type x, as the sum of all the remaining stocks of DEHP for products of type x placed on the market between year n and the year (n the lifespan of products of type x). After being used, i.e. present on the market, products and stocks of DEHP contained in products are either incinerated or placed in landfill and therefore leave the stock of DEHP present on the market.

$$\begin{aligned} &Additional Stock of DEHP_{(article x) (year N)} \\ &= Quantity of DEHP marketed_{article x (year N)} \\ &+ Quantity of DEHP recycled_{article x (year N)} \end{aligned} \quad (2)$$

$$\begin{aligned}
& \text{Remaining Stock of DEHP}_{(article\ x)(year\ N, year\ n)} = \text{Stock of DEHP}_{(article\ x)(year\ N, year\ n-1)} - \\
& \text{Emissions from use}_{(article\ x)(year\ n-1)} = \text{Stock of DEHP}_{(article\ x)(year\ N)} - \\
& \sum_{i=N}^{i=n} \text{Emission from use}_{(article\ x)_{year\ (i)}} \quad (3)
\end{aligned}$$

$$\begin{aligned}
& \text{Total Stock DEHP}_{article\ x\ (year\ n)} \\
& = \sum_{i=n - \text{life span of the article } x + 1}^{i=n} \text{Remaining Stock DEHP}_{(article\ x)(year\ N=i, year\ n)} \quad (4)
\end{aligned}$$

We have assumed that European DEHP production began in 1945, notably to produce wires and cables (The European Council of Vinyl Manufacturers). According to the data sources consulted, there is no information on DEHP production and on quantities used by the European industry between 1945 and 1993. To fill this data gap, we assumed that the growth of DEHP production and uses have the same exponential growth as shown by the available PVC production data (Purser D. 2015). After the year 1997, more data are available in the literature (European-Chemicals-Bureau 2008, Lindeboom R. et al. 2009, COWI 2013) but not for every year of the period studied. To fill missing years, linear interpolation has been carried out. The scarcity of data for uses of DEHP before 1997 contributes to the uncertainty of the estimation of stocks. More information regarding the method and data used are presented in Suppl mat – Table 1, Table 2 and Table 3.

As indicated above, the possible dislocation of products during all their service life and subsequent releases of “pieces” of products which then emit a significant amount of DEHP into the environment has been considered for outdoor products (garden products, plastics used in agriculture...) but not for indoor products, because it might be significant due to a lack of data for emission factors in the literature. Emissions of DEHP from pieces released from used and broken products have been estimated following Eq.5. EFs are applied for all the lifespan of the product and are presented in Table 9 in the supplementary material.

$$\begin{aligned}
& \text{Emission due to dislocation of outdoor article (ED)}_{article\ (x)_{year\ (N)}} \\
& = \text{Quantity of DEHP marketed}_{article\ x\ (year\ N)} \\
& * \text{EF dislocation}_{article\ (x)} \quad (5)
\end{aligned}$$

Finally, different specific emission factors are associated to each stage of each product’s life cycle, and to each environmental compartment. They are expressed in mass percentage of DEHP emitted in relation to the amount of DEHP present in the product, or of DEHP manufactured, or of DEHP treated at end-of life. Environmental compartments considered in this study are air, water and soil. Factors used for these estimations have been selected from (European-Chemicals-Bureau 2008, Lindeboom R. et al. 2009, COWI 2013). In the literature used for the definition of emission factors, emissions to water are distinguished between water emissions reaching wastewater treatment plants and stormwater which does not reach wastewater treatment plants. We assumed no removal of DEHP in wastewater treatment plants (WWTPs) since no overall removal of this compound in WWTPs at a European scale can be easily assessed. This is a conservative approach, overestimating emissions through the water compartment. If removal in WWTP had been considered, it would anyway have led to the need to consider transfers of DEHP from wastewater to soil via WWTP sludge, for which there is little knowledge available. As no water treatment has been considered, emissions to wastewater and stormwater have been summed up and grouped together under the water compartment. Another assumption overestimating emissions of DEHP for all the compartments is the non-consideration of degradation of the substance in products and in the environment.

Treatment of wastes and leachates in landfill changes during the landfill life (exploitation and post-exploitation) and varies from a landfill to another. We assumed that landfills could emit DEHP for up to 100 years, based on the opinions of in-house Ineris experts who took into account the time PVC products fragments could remain able to emit DEHP. This assumption has been based on (COWI 2013) and (Defranceschi M. 2015). As a sensitivity analysis, we also have carried out a calculation considering 30 years for DEHP emissions from landfill.

Table 2 Emission factors

DEHP or products life cycle step	Emission factors	Reference
DEHP production	EF air, EF soil, EF wastewater	(European-Chemicals-Bureau 2008)
Manufacture of film/sheet and coated product and roofing calendered	EF air, EF wastewater	(European-Chemicals-Bureau 2008)
Manufacture of wire and cable, hoses and profiles	EF air, EF wastewater	(European-Chemicals-Bureau 2008)
Manufacture of moulded products	EF air, EF wastewater	(European-Chemicals-Bureau 2008)
Manufacture of medical device	EF air, EF wastewater	(European-Chemicals-Bureau 2008)
Manufacture of roofing coil coated, flooring and wall covering	EF air, EF wastewater	(European-Chemicals-Bureau 2008)
Manufacture of “car undercoating”	EF air, EF wastewater	(European-Chemicals-Bureau 2008)
Manufacture of adhesive and sealant, lacquers and paints, printing ink, ceramic products	EF air, EF soil, EF wastewater	(European-Chemicals-Bureau 2008)
Indoor uses of all products except flooring, adhesive and sealant, lacquers and paints, printing ink, ceramic products	EF air	(European-Chemicals-Bureau 2008)
Flooring products uses	EF air, EF wastewater	(European-Chemicals-Bureau 2008)
All outdoor uses except adhesive and sealant, lacquers and paints, printing ink, ceramic products	EF air, EF soil, EF stormwater	(European-Chemicals-Bureau 2008)
Indoor use of adhesive and sealant, lacquers and paints, printing ink and ceramic products	EF air, EF wastewater	(European-Chemicals-Bureau 2008)
Outdoor use of adhesive and sealant, lacquers and paints, printing ink, ceramic products	EF air, EF stormwater	(European-Chemicals-Bureau 2008)
Dismantling of products (all types) during outdoor uses	EF air, EF stormwater, EF soil	(European-Chemicals-Bureau 2008)

Incineration of products (all types)	EF air	(European-Chemicals-Bureau 2008)
Landfilling of products (all types)	EF soil	Hypothesis based on (COWI 2013) and (Defranceschi M. 2015)
Recycling of products (all types)	No data available in the literature	
Shredding of car undercoating	EF air, EF soil	(COWI 2013)

2.2 DEFINITION OF REGULATION AND RECYCLING SCENARIOS

We used 1945 as the start of DEHP production and use in Europe and our scenarios first reconstitute the annual emissions since 1945 to 2020 and forecast future annual emission from 2020 until 2040, under several future scenario assumptions.

We first define a future **baseline scenario** in which after 2020, DEHP quantities in new products are based on the European Chemicals Agency (ECHA)'s business-as-usual predictions in the absence of the latest market restrictions that have entered into force in 2020 (ECHA 2016). This baseline scenario is used as a comparison basis for other scenarios and to illustrate the potential very long-term emissions of DEHP to the environment caused by its accumulated stock in the economy since 1945. (ECHA 2016, European Commission 2018).

We define another future “**restrictive market regulation scenario**”, in order to assess the impact of the recent restriction under REACH prohibiting after 2020 the production and the importation and placing on the market of products containing DEHP for indoor use and/or which may have significant skin contact (European Commission 2018). In this scenario the total quantity of DEHP in products placed on the European market is reduced by 70% compared to the baseline scenario in 2020 and by 75% in 2040.

Another aim of our work was also to assess the impact of the prohibition by REACH to recycle PVC products without removing DEHP, on future DEHP emissions. In fact, DEHP contained in PVC products represent an important part of the total stock of DEHP contained in products. Technologies to remove DEHP during recycling of PVC products are under development but not yet available (Wagner S., 2020). Therefore, we compared the future situation (no further recycling without DEHP removal, that is, in practice no recycling of PVC products containing DEHP) to three “**PVC recycling scenarios**”, by varying the distribution of end-of-life treatments between incineration, landfilling and recycling:

- A first PVC recycling scenario called “moderate recycling” with a level of recycling set at 26% in 2015 based on (European Commission 2018). We assume 35% in 2030 with 2% growth per year between 2015 and 2030.
- A second PVC recycling scenario called “high recycling” with 26% in 2015 based on (European Commission 2018). We assume 60% in 2030 with 3% growth per year between 2015 and 2030.
- For these two latter scenarios the percentage of products recycled is stable over the period of 2030 2040, and it has been considered that the additional recycling is replacing landfilling.

We considered under these recycling scenarios that quantities of DEHP recycled with the PVC material are added to the quantities already present on the market. Since it is unclear from data sources whether the data we use for products placed on the market already take some account of recycling, there is a possibility for some double counting of DEHP placed on the market from recycling.

More information on data and assumption used for the development of scenarios are presented in the supplementary material, in particular Suppl mat – Table 1 and Table 2.

3 RESULTS AND DISCUSSION

3.1 BASELINE AND IMPACTS OF REGULATION

In this section, the impact of regulatory restrictions and the impact of prohibiting recycling of DEHP within PVC, on stocks and on emissions to all environmental media, are presented. It should be reminded that given the scarcity of data and because of all the assumptions that were needed, uncertainties are high.

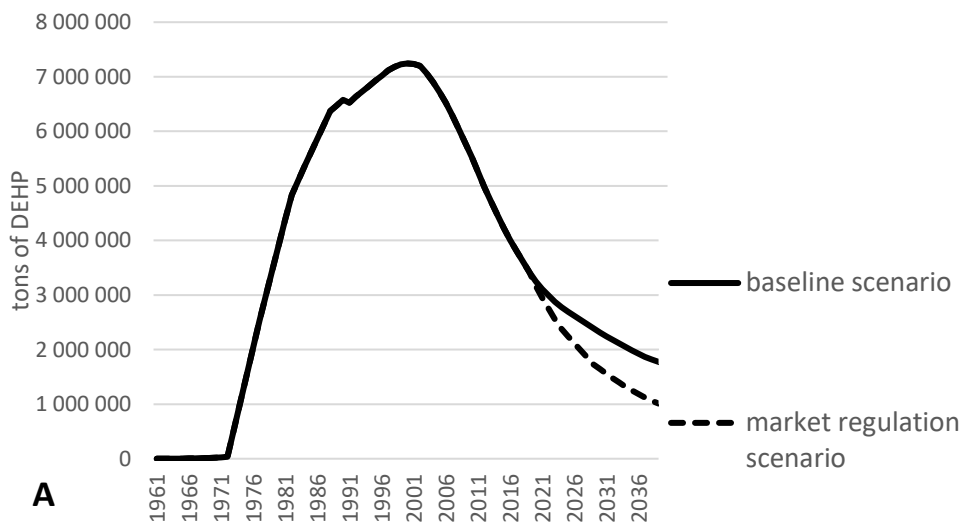


Fig. 2 Comparison of past and future DEHP stocks for the “restrictive market regulation” and for the “baseline” scenarios

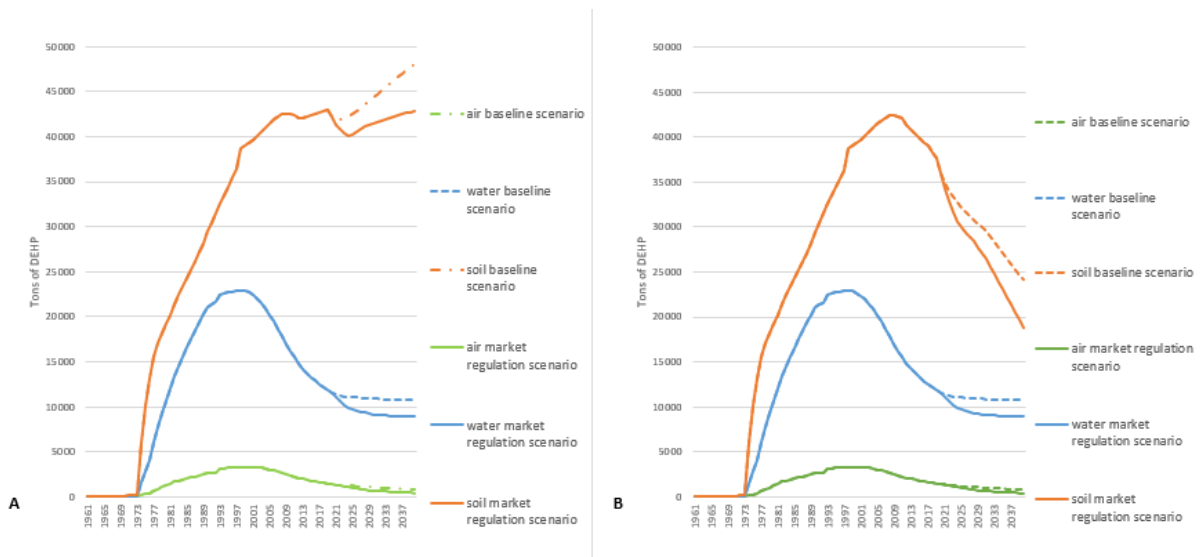


Fig. 3 Comparison of DEHP emissions per environmental compartment with or without restrictive market regulation with the assumption of 100 lifetime years in landfill (A) and with the assumption of 30 lifetime years in landfill (B)

The peak of total emissions is reached around 2001, with or without market restriction and for the two assumptions of lifetime in landfill and corresponds with a time lag of 2 years to the peak of DEHP contained in products used in Europe, reached in 2000, as illustrated in Fig. 2. This result is consistent with trends of phthalates concentrations in European surface waters reported in the review by (Bergé A. et al., 2013) that also exhibit a maximum around year 2000. It is also consistent with DEHP stocks estimated in the EU by the (European Commission 2018) and for Japan in (Muchangos L.D., Xue M. et al. 2019).

Figure 3 illustrates emissions of DEHP to all environmental compartments (water, air, soil) for all steps of the chemical substance's life cycle, over the period of 1960-2040. Results are shown for the two assumptions regarding the time spent by used products in landfills (A = 100 years and B = 30 years). Total emissions are strongly dominated by emissions to soil emissions to air appear marginal.

Considering the baseline scenario, even if the industry has significantly switched to alternatives to DEHP, emissions decrease is slow, and high emissions are to be expected well beyond 2040 for water and soil under the assumption of 100 years lifetime of products in landfills. On the opposite, if we consider a shorter lifetime (30 years), DEHP emissions to soil follow a strong decrease after 2010, but still would be significant in 2040.

DEHP, despite not being considered and prioritized as a persistent chemical from a regulatory perspective, has been emitted in the EU at a high level for at least 50 years. The restrictive market regulation scenario that is forbidding most remaining uses of DEHP appears to have only limited impacts on both stocks and emissions after 2011, the year of implementation of the regulation. It appears to be too late for market regulation to offset the combined effects of past stocks buildup and of landfilling.

3.2 IMPACTS OF RECYCLING ON THE EMISSIONS OF DEHP

Logically, the prohibition of recycling accelerates the reduction of DEHP stocks on the market (Fig. 4A), and therefore, the same influence of prohibiting recycling of DEHP within PVC is seen regarding emissions in landfills (Fig. 4B) and in total emissions (Fig. 5).

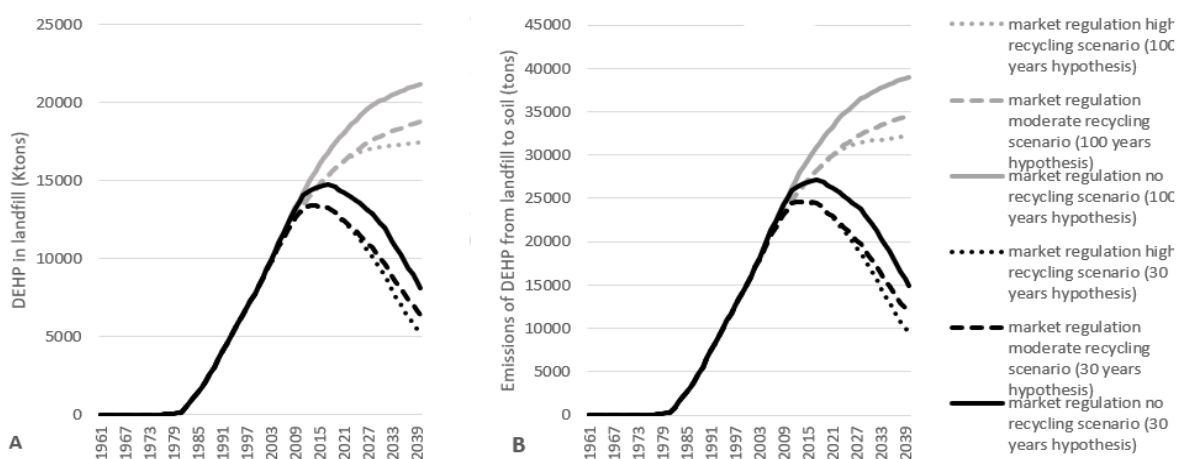


Fig. 4 Past and future stocks of DEHP in landfill (A) and emissions of DEHP from landfills through the soil compartment (B)

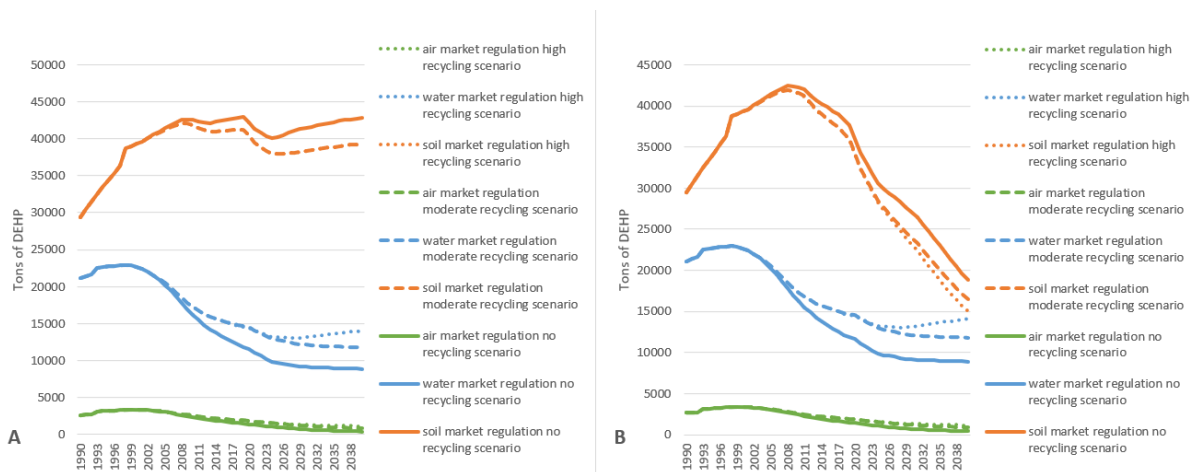


Fig. 5 Impacts of the prohibition of recycling DEHP in PVC on DEHP emissions, with 100 years lifetime in landfill (A) with 30 years lifetime in landfill (B)

Overall, despite recycling having an influence on both stocks and emissions, the duration of emissions during landfilling is the most important driver for future stock (Fig.4A) and future emission trends (Figs 4B and 5), especially regarding emissions to soil which are dominating total emissions.

Emission to soil from landfill could increase by a factor of 8 between 1990 and 2040 (Fig 4B) in the scenario without recycling and by a factor of 6.5 in the scenario with recycling.

In all scenarios, emissions to water and air at best decrease very slowly, especially when higher recycling rates are chosen. Technologies that remove DEHP from products as, for example, PVC products prior to recycling could allow recycling in the future, but their feasibility and sustainability is still to be confirmed (Wagner et al., 2020).

4 CONCLUSIONS

In this study, we developed retrospective and prospective scenarios to assess past and future emissions of DEHP in the environment, taking into account the entire lifecycle of the substance, from its production and its inclusion in products to the recycling and end of life of these products. Our results tend to show that the introduction of recent regulations to prohibit 75% of the use of DEHP or prohibit its recycling within PVC products do not further reduce significantly future emissions (reduction of only 10% and 13%, respectively, in 2030 and in 2040 compared to the baseline scenario).

This long-term persistence of emissions is explained by several causes: the inertia of the industrial and risk regulation systems that let high stocks build in the economy and the long-term presence of waste in landfills. Despite DEHP not being an environmental persistent pollutant, these simulations suggest it will remain a cause of environmental contamination many decades after uses have declined and even ceased, notwithstanding stocks that may have also built in the soil environment downstream of environmental emissions. It is likely that several chemicals that are not considered as persistent chemicals and therefore not the focus of international regulations could exhibit the same characteristics and, in addition, as DEHP, be transported over long distances within exported and imported products. Another aspect not considered in the study is the degradation of DEHP in the environment and the long-term release of DEHP stocked in the soil compartment from other environmental compartments. These factors should probably reduce the amount of emissions per year but further increase the long-term emission (Muchangos L.D., Xue M. et al. 2019)(Briand C., 2016).

These facts suggest that current regulatory procedures to grant or withdraw access of chemicals to the market who do take into account the intrinsic properties of chemicals that make them chemically persistent or susceptible of long-range transport (Stockholm Convention, REACH Regulation) should consider also integrating use patterns that make chemicals persistent and susceptible of long-term

presence and long-range transport in products, because these properties have the potential to create long-term and almost irreversible environmental pollution and impacts.

Supplementary information

Author contribution

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Valentin Chapon and Jean-Marc Brignon. The first draft of the manuscript was written by Valentin Chapon and Jean-Marc Brignon. All authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials

On request the authors can transmit all data used and estimations carried out for this work.

Declaration

Consent to participate

“Not applicable” because this work doesn’t involve anybody except the authors.

Consent for the publication

“Not applicable” because no individual data have been used for this study.

Competing interests

The authors declare no competing interests.

References

- Bergé A, Cladière M, Gasperi J, Coursimault A, Tassin B, Moilleron R (2013) Meta-analysis of environmental contamination by phthalates. *Environ Sci Pollut Res Int* 20(11):8057–76. <https://doi.org/10.1007/s11356-013-1982-5>
- Bonnard M., et al. (2020). "Experience Gained from Ecotoxicological Studies in the Seine River and Its Drainage Basin Over the Last Decade: Applicative Examples and Research Perspectives." *Springer* **90**.
- Brignon J.M. (2021). "Costs and benefits of recycling PVC contaminated with the legacy hazardous plasticizer DEHP." *Waste Manag Res* **39**(9): 1185-1192.
- Brignon J.M., et al. (2018). PIREN-Seine phase VII - rapport 2018 – Estimation des émissions de Déca-BDE sur le bassin de la Seine
- COWI (2009). Data on manufacture, import, export, uses and releases of bis(2-ethylhexyl)phthalate (dehp) as well as information on potential alternatives to its use
- COWI (2013). Hazardous substances in plastic materials.
- Defranceschi M., et al. (2015). Substances émergentes, polluants émergents dans les déchets : Le cas des phtalates. *RECORD*, RECORD. **ETUDE N° 13-0151/1A**
- Earnshaw M.R., et al. (2013). "Estimating European historical production, consumption and atmospheric emissions of decabromodiphenyl ether." *Sci Total Environ* **447**: 133-142.
- ECHA (2008). Member state committee support document for identification of Bis(2-ethylhexyl)phthalate (DEHP) as a substance of very high concern.
- ECHA (2016). Trade in phtalates excel files for the dossier development of the SEA. ECHA, ECHA.
- European-Chemicals-Bureau (2008). European Union Risk Assessment Report : bis(2-ethylhexyl)phthalate (DEHP).
- European Commission (2018). "RÈGLEMENT (UE) 2018/2005 DE LA COMMISSION du 17 décembre 2018 modifiant l'annexe XVII du règlement (CE) no 1907/2006 du Parlement européen et du Conseil concernant l'enregistrement, l'évaluation et l'autorisation des substances chimiques, ainsi que les restrictions applicables à ces substances (REACH), en ce qui concerne le phtalate de bis(2-éthylhexyle) (DEHP), le phtalate de dibutyle (DBP), le phtalate de benzyle et de butyle (BBP) et le phtalate de diisobutyle (DIBP)." *Journal officiel de l'Union européenne*.
- European Commission (2020). COMMISSION STAFF WORKING DOCUMENT FITNESS CHECK on endocrine disruptors.
- Gasperi J., et al. (2018). "Contamination of soils by metals and organic micropollutants: case study of the Parisian conurbation." *Environ Sci Pollut Res Int* **25**(24): 23559-23573.
- IARC (2013). Some Chemicals Present in Industrial and Consumer Products, Food and Drinking-water IARC Monographs on the Evaluation of Carcinogenic Risks to Humans Volume 101.
- Kleijn R., et al. (2000). "Dynamic substance flow analysis: the delaying mechanism of stocks, with the case of PVC in Sweden." *Ecological Economics* **32**: 241-254.
- Lindeboom R., et al. (2009). Source Control of Priority Substances in Europe An Inventory and Assessment of Options for Reducing Emissions: DEHP.
- Muchangos L.D., et al. (2019). "Flows, stocks, and emissions of DEHP products in Japan." *Sci Total Environ* **650**(Pt 1): 1007-1018.
- Norwegian Institute for Air Research (2007). SOCOPSE Workpackage 3 – D3.1.
- Purser D. (2015). *Toxicology, survival and health hazards of combustion products (chapter 12)*.
- Song R., et al. (2017). "Dynamic Model for the Stocks and Release Flows of Engineered Nanomaterials." *Environ Sci Technol* **51**(21): 12424-12433.
- Teil M.J., et al. (2007). "Occurrence of phthalate diesters in rivers of the Paris district (France)." *hydrological processes* **21**.
- The European Council of Vinyl Manufacturers. "www.pvc.org." Retrieved june, 2017, from http://www.pvc.org/en/search_engine/pvc%20world%20production%201950-2000.
- Wagner S., et al. (2020). "Legacy additives in a circular economy of plastics: Current dilemma, policy analysis, and emerging countermeasures." *Resources, Conservation and Recycling* **158**.

Wikipédia. "PVC clothing." 2021, from https://en.wikipedia.org/wiki/PVC_clothing.

SUPPLEMENTARY MATERIAL

- 1) References and assumptions used for the estimations of DEHP produced, manufactured, and marketed

Supp mat – Table 1: References and data used for the estimation of quantities of DEHP produced in Europe (tons of DEHP) [extract]

Years	1945	1993	1997	2004	2007	2008 - 2009	2010 - 2040
Quantity of DEHP (tons) produced in Europe	1	595,000 (Norwegian Institute for Air Research 2007)	595,000 (Norwegian Institute for Air Research 2007)	247,000 (Norwegian Institute for Air Research 2007)	100,000 (Norwegian Institute for Air Research 2007)	No data for this period estimation for each year with linear interpolation between 2007 and 2010	0

All data used for the estimation of DEHP produced in Europe are presented in Supp mat-Table 1. To fill the data gap between 1945 and 1993, we assumed that the growth of DEHP production in Europe has had the same exponential growth as PVC production (Purser D., 2015). Based on this assumption an exponential interpolation (Eq. 6 below), has been used to estimate quantities of DEHP produced during this period.

Eq. 6

$$\text{Quantity of DEHP produced in Europe (year } n) = 0,8487 * e^{0,1946 * ((\text{year } n) - 1944)} \quad (6)$$

After 1993 linear projection have been used to estimate quantities produced for the years for which no data exist in the literature (from 1994 to 1996, from 1998 to 2003, for 2005, 2006, 2008 and 2009).

For the years 2010 to 2040 no production of DEHP in Europe has been considered. According to (European Commission, 2020) European production of DEHP strongly decline between 2005 and 2013 and only one producer of DEHP exists in 2020.

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Supp mat – Table 2: References and data used for the estimation of quantities of DEHP used for the manufacture of products (tons of DEHP)

All data used for the estimation of DEHP used in Europe for the manufacture of all products are presented in Supp Mat-Table 2.

Years	1945	1993	1997	2007	2011	2012 - 2019	2020	2021 - 2039	2040
Quantity of DEHP used by industry to manufacture products	1	476,000 (European-Chemicals-Bureau , 2008)	533,325 (Norwegian Institute for Air Research 2007)	282,120 (Norwegian Institute for Air Research 2007)	107,526 (ECHA 2016) data for the following products : flooring, wall covering, film/sheet and coated products, wires and cables, hoses and profiles, coated fabric indoor use, moulded products Authors own assumption of 1% decrease per year since 2007 for the following products: car undercoating, Roofing calendered & coil coated, coated fabric outdoor use, adhesive and scellant, lacquers and paints, printing ink and ceramic	(ECHA 2016) data for the following products : flooring, wall covering, film/sheet and coated products, wires and cables, hoses and profiles, coated fabric indoor use, moulded products Authors own assumption of 1% decrease per year since 2007 for the following products: car undercoating, Roofing calendered & coil coated, coated fabric outdoor use, adhesive and scellant, lacquers and paints, printing ink and ceramic	<u>Baseline scenario</u> 44,902 (ECHA 2016) data for the following products : flooring, wall covering, film/sheet and coated products, wires and cables, hoses and profiles, coated fabric indoor use, moulded products Authors own assumption of 1% decrease per year since 2007 for the following products: car undercoating, Roofing calendered & coil coated, coated fabric outdoor use, adhesive and scellant, lacquers and paints, printing ink and ceramic	<u>Baseline scenario</u> ... (ECHA 2016) data for the following products : flooring, wall covering, film/sheet and coated products, wires and cables, hoses and profiles, coated fabric indoor use, moulded products Authors own assumption of 1% decrease per year since 2007 for the following products: car undercoating, Roofing calendered & coil coated, coated fabric outdoor use, adhesive and scellant, lacquers and paints, printing ink and ceramic	<u>Baseline scenario</u> 33,263 (ECHA 2016) data for the following products : flooring, wall covering, film/sheet and coated products, wires and cables, hoses and profiles, coated fabric indoor use, moulded products Authors own assumption of 1% decrease per year since 2007 for the following products: car undercoating, Roofing calendered & coil coated, coated fabric outdoor use, adhesive and scellant, lacquers and paints, printing ink and ceramic
							<u>Regulation scenario</u> 31,074 (ECHA 2016) data for the following products : flooring, wall covering, film/sheet and coated products, wires and cables, hoses and profiles, coated fabric indoor use, moulded products	<u>Regulation scenario</u> ... (ECHA 2016) data for the following products : flooring, wall covering, film/sheet and coated products, wires and cables, hoses and profiles, coated fabric indoor use, moulded products	<u>Regulation scenario</u> 23,705 (ECHA 2016) data for the following products : flooring, wall covering, film/sheet and coated products, wires and cables, hoses and profiles, coated fabric indoor use, moulded products

							Authors own assumption of 1% decrease per year since 2007 for the following products: car undercoating, Roofing calendered & coil coated, coated fabric outdoor use, adhesive and scelant, lacquers and paints, printing ink and ceramic	Authors own assumption of 1% decrease per year since 2007 for the following products: car undercoating, Roofing calendered & coil coated, coated fabric outdoor use, adhesive and scelant, lacquers and paints, printing ink and ceramic	Authors own assumption of 1% decrease per year since 2007 for the following products: car undercoating, Roofing calendered & coil coated, coated fabric outdoor use, adhesive and scelant, lacquers and paints, printing ink and ceramic
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6 To fill the data gap between 1945 and 1993, we assumed that the growth of DEHP quantities used for the manufacture of products in Europe have the same
7 exponential growth as PVC production (Purser D. 2015). Based on this assumption we used an exponential interpolation, based on equation n°7.

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9 **Eq. 7**

10 $Quantity\ of\ DEHP\ produced\ in\ Europe_{(year\ n)} = 0,8471 * e^{0,1915 * ((year\ n) - 1944)} \quad (7)$

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12 After 1993 linear interpolations have been used to estimate quantities manufactured for the years for which no data exist in the literature (from 1994 to 1996,
13 from 1998 to 2006 and from 2008 to 2010).

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15 All the references presented in the table 4 provide data of DEHP used for the manufacture of products per type of products except (ECHA 2016) which not
16 provide information for car undercoating, roofing calendered & coil coated products, coated fabric outdoor used, adhesive and sealant, lacquers and paints,
17 printing ink and ceramic. For these products from 2007 to 2040 linear extrapolation with 1% decrease per year has been considered.

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Supp mat – Table 3: References and data used for the estimation of quantities of DEHP contained in products placed on the market in Europe between 1960 and 2040 per type of product (tons of DEHP)

Years	1945	1960	1965	1973	1975	1997	2007	2011	2020	2040
Quantity of DEHP contained in products placed in the market										
Total quantity of DEHP contained in products placed on the market used for the estimation of stocks	1	No data for this year	No data for this year	500,000 (Norwegian Institute for Air Research 2007)	No data for this year	538,761 (Norwegian Institute for Air Research 2007)	282,420 (Norwegian Institute for Air Research 2007)	No data for this year	No data for this year	No data for this year
Flooring	Not used before 1975					73,780 (Norwegian Institute for Air Research 2007)	32,200 (Norwegian Institute for Air Research 2007)	22,611 (ECHA 2016)	<u>Baseline scenario</u> 28,013 (ECHA 2016)	<u>Baseline scenario</u> 34,207 (ECHA 2016)
									<u>Regulation scenario</u> 28,013 (ECHA 2016)	<u>Regulation scenario</u> 34,207 (ECHA 2016)
Wall covering	Not used before 1975					98,780 (Norwegian Institute for Air Research 2007)	10,100 (Norwegian Institute for Air Research 2007)	7092 (ECHA 2016)	<u>Baseline scenario</u> 8787 (ECHA 2016)	<u>Baseline scenario</u> 10,730 (ECHA 2016)
									<u>Regulation scenario</u> 0 (ECHA 2016)	<u>Regulation scenario</u> 0 (ECHA 2016)
Coated fabric indoor use	Not used before 1960	No data for this period				21,000 (Norwegian Institute for Air Research 2007)	31,800 (Norwegian Institute for Air Research 2007)	16,117 (ECHA 2016)	<u>Baseline scenario</u> 12,143 (ECHA 2016)	<u>Baseline scenario</u> 13,782 (ECHA 2016)

						<u>Regulation scenario</u> 0 (ECHA 2016)	<u>Regulation scenario</u> 0 (ECHA 2016)
Coated fabric outdoor use			52,171 (Norwegian Institute for Air Research 2007)	12,800 (Norwegian Institute for Air Research 2007)	12,296 (ECHA 2016)	<u>Baseline scenario</u> 11,232 (ECHA 2016)	<u>Baseline scenario</u> 9187 (ECHA 2016)
						<u>Regulation scenario</u> 11,232 (ECHA 2016)	<u>Regulation scenario</u> 9187 (ECHA 2016)
Film/sheet & coated products	Not used before 1960	No data for this period	71,400 (Norwegian Institute for Air Research 2007)	41,200 (Norwegian Institute for Air Research 2007)	25,107 (ECHA 2016)	<u>Baseline scenario</u> 19,551 (ECHA 2016)	<u>Baseline scenario</u> 23,193 (ECHA 2016)
						<u>Regulation scenario</u> (ECHA 2016) 0	<u>Regulation scenario</u> (ECHA 2016) 0
Wires and cables indoor use	Not used before 1965	No data for this period	60,920 (Norwegian Institute for Air Research 2007)	52,600 (Norwegian Institute for Air Research 2007)	32,441 (ECHA 2016)	<u>Baseline scenario</u> 0 (ECHA 2016)	<u>Baseline scenario</u> 0 (ECHA 2016)
						<u>Regulation scenario</u> (ECHA 2016)	<u>Regulation scenario</u> (ECHA 2016)

Wires and cables outdoor use			20,000 (Norwegian Institute for Air Research 2007)	12,100 (Norwegian Institute for Air Research 2007)	6067 (ECHA 2016)	<u>Baseline scenario</u> 0 (ECHA 2016)	<u>Baseline scenario</u> 0 (ECHA 2016)
			<u>Regulation scenario</u> 0 (ECHA 2016)	<u>Regulation scenario</u> 0 (ECHA 2016)			
Moulded products indoor use	Not used before 1960	No data for this period	10,309 (Norwegian Institute for Air Research 2007)	5000 (Norwegian Institute for Air Research 2007)	7976 (ECHA 2016)	<u>Baseline scenario</u> 8028 (ECHA 2016)	<u>Baseline scenario</u> 9618 (ECHA 2016)
			<u>Regulation scenario</u> 0 (ECHA 2016)	<u>Regulation scenario</u> 0 (ECHA 2016)			
Moulded products outdoor use			40,000 (Norwegian Institute for Air Research 2007)	19,400 (Norwegian Institute for Air Research 2007)	30,946 (ECHA 2016)	<u>Baseline scenario</u> 31,150 (ECHA 2016)	<u>Baseline scenario</u> 37,320 (ECHA 2016)
			<u>Regulation scenario</u> 0 (ECHA 2016)	<u>Regulation scenario</u> 0 (ECHA 2016)			
Roofing calendered & coil coated	Not used before 1960	No data for this period	6000 (Norwegian Institute for Air Research 2007)	3600 (Norwegian Institute for Air Research 2007)	3458 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 3159 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 2584 (Authors own assumption 1% decrease per year since 2007)
			<u>Regulation scenario</u> 3159	<u>Regulation scenario</u> 431			

						(Authors own assumption 1% decrease per year since 2007)	(Authors own assumption 1% decrease per year since 2007)
Hoses and profiles indoor use	Not used before 1960	No data for this period	57,120 (Norwegian Institute for Air Research 2007)	29,600 (Norwegian Institute for Air Research 2007)	14,651 (Norwegian Institute for Air Research 2007)	<u>Baseline scenario</u> 5338 (ECHA 2016)	<u>Baseline scenario</u> 3400 (ECHA 2016)
						<u>Regulation scenario</u> 5338 (ECHA 2016)	<u>Regulation scenario</u> 3400 (ECHA 2016)
Hoses and profiles outdoor use	Not used before 1960	No data for this period	6000 (Norwegian Institute for Air Research 2007)	3700 (Norwegian Institute for Air Research 2007)	1831 (ECHA 2016)	<u>Baseline scenario</u> 667 (ECHA 2016)	<u>Baseline scenario</u> 425 (ECHA 2016)
						<u>Regulation scenario</u> 667 (ECHA 2016)	<u>Regulation scenario</u> 425 (ECHA 2016)
Car undercoating	Not used before 1960	No data for this period	7000 (Norwegian Institute for Air Research 2007)	4000 (Norwegian Institute for Air Research 2007)	3842 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 3510 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 2871 (Authors own assumption 1% decrease per year since 2007)
						<u>Regulation scenario</u> 3510 (Authors own assumption 1% decrease per year since 2007)	<u>Regulation scenario</u> 2870 (Authors own assumption 1% decrease per year since 2007)
Adhesive and sealant indoor use			6105	6105	3842	<u>Baseline scenario</u>	<u>Baseline scenario</u>

			(Norwegian Institute for Air Research 2007)	(Norwegian Institute for Air Research 2007)	(Authors own assumption 1% decrease per year since 2007)	3510 (Authors own assumption 1% decrease per year since 2007)	2871 (Authors own assumption 1% decrease per year since 2007)
						<u>Regulation scenario</u> 3510 (Authors own assumption 1% decrease per year since 2007)	<u>Regulation scenario</u> 2871 (Authors own assumption 1% decrease per year since 2007)
Adhesive and sealant outdoor use	Not used before 1960	No data for this period	5036 (Norwegian Institute for Air Research 2007)	3300 (Norwegian Institute for Air Research 2007)	3169 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 2896 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 2369 (Authors own assumption 1% decrease per year since 2007)
						<u>Regulation scenario</u> 2896 (Authors own assumption 1% decrease per year since 2007)	<u>Regulation scenario</u> 2369 (Authors own assumption 1% decrease per year since 2007)
Lacquers and paints total indoor use	Not used before 1960	No data for this period	804 (Norwegian Institute for Air Research 2007)	500 (Norwegian Institute for Air Research 2007)	480 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 439 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 359 (Authors own assumption 1% decrease per year since 2007)
						<u>Regulation scenario</u> 351 (Authors own assumption 1% decrease per year since 2007)	<u>Regulation scenario</u> 287 (Authors own assumption 1% decrease per year since 2007)
Lacquers and paints total outdoor use			644 (Norwegian Institute for Air Research 2007)	400 (Norwegian Institute for Air Research 2007)	384	<u>Baseline scenario</u> 439	<u>Baseline scenario</u> 359 (Authors own assumption 1% decrease per year since 2007)

					(Authors own assumption 1% decrease per year since 2007)	(Authors own assumption 1% decrease per year since 2007)	
						<u>Regulation scenario</u> 351 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 287 (Authors own assumption 1% decrease per year since 2007)
printing ink	Not used before 1960	No data for this period	1661 (Norwegian Institute for Air Research 2007)	1000 (Norwegian Institute for Air Research 2007)	961 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 878 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 718 (Authors own assumption 1% decrease per year since 2007)
						<u>Regulation scenario</u> 878 (Authors own assumption 1% decrease per year since 2007)	<u>Regulation scenario</u> 718 (Authors own assumption 1% decrease per year since 2007)
Ceramic	Not used before 1960	No data for this period	29 (Norwegian Institute for Air Research 2007)	20 (Norwegian Institute for Air Research 2007)	19 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 18 (Authors own assumption 1% decrease per year since 2007)	<u>Baseline scenario</u> 15 (Authors own assumption 1% decrease per year since 2007)
						<u>Regulation scenario</u> 18 (Authors own assumption 1% decrease per year since 2007)	<u>Regulation scenario</u> 15 (Authors own assumption 1% decrease per year since 2007)

All data used for the estimation of DEHP contained in products placed on the European market are presented in the Supp – Mat 3. We consider that no product has been placed on the market before 1960. No data for DEHP contained in products placed on the European market for each type of product exists before 1997. Quantities of DEHP contained in each type of product placed on the European market has been estimated in two steps:

- First, quantities of DEHP contained in all the products placed on the European market between 1960 and 1997 have been estimated with the equation n°8. As for quantities of DEHP produced and manufactured, an exponential growth of DEHP placed in the market has been considered (Purser D., 2015)

Eq. 8

$$\text{Quantity of DEHP marketed}_{(year N)} = 0,7216 * e^{0,3319 * ((year N) - 1944)} \quad (8)$$

- Second, by using data of (Norwegian Institute for Air Research 2007) for the year 1997, the DEHP placed on the market per type of product has been estimated using the equation n° 9:

Eq. 9

$$\text{Quantity of DEHP marketed}_{(product x)_{(year N)}} = \frac{\text{Quantity of DEHP marketed}_{(product x)_{(1997)}}}{\text{Total Quantity of DEHP marketed}_{1997}} * \text{Total Quantity of DEHP marketed}_{(year N)} \quad (9)$$

After 1997, linear interpolation has been used for the years: 1998 to 2006 and 2008 to 2010. After 2011, data from ECHA has been used for the following products: flooring, wall covering, coated fabric, film/sheet & coated products, wires and cables, moulded products, roofing calendered & coil coated products, hoses and profiles. For the other products, which are not targeted by the European regulation a 1% decrease per year after 2007 has been assumed.

2) EMISSIONS FACTORS

Supp mat – Table 4: Emission Factors for production of DEHP

	Emission Factors expressed in percentages of DEHP emitted as percentages of weight of DEHP produced		Reference
	Environmental compartment	Value	
DEHP production	EF air	0.0003%	(European-Chemicals-Bureau 2008)
	EF soil	0.001%	
	EF wastewater	0.06%	

Supp mat – Table 5: Emission factors for the manufacture products

Manufacture of products	Emission Factors expressed in percentages of DEHP used for the production of product		References
	Environmental compartment	Value	
Manufacture of flooring	EF air	0.09%	(European-Chemicals-Bureau 2008)
	EF wastewater	0.09%	
	EF soil	0.00%	
Manufacture of wall covering	EF air	0.09%	(European-Chemicals-Bureau 2008)
	EF wastewater	0.09%	
	EF soil	0.00%	
Manufacture of indoor coated fabric	EF air	0.11%	(European-Chemicals-Bureau 2008)
	EF wastewater	0.11%	
	EF soil	0.00%	
Manufacture of outdoor coated fabric	EF air	0.09%	(European-Chemicals-Bureau 2008)
	EF wastewater	0.09%	
	EF soil	0.00%	
Manufacture of film/sheet & coated product	EF air	0.03%	(European-Chemicals-Bureau 2008)
	EF wastewater	0.09%	
	EF soil	0.00%	
Manufacture of wires and cables	EF air	0.01%	(European-Chemicals-Bureau 2008)
	EF wastewater	0.01%	
	EF soil	0.00%	
Manufacture of moulded product	EF air	0.01%	(European-Chemicals-Bureau 2008)
	EF wastewater	0.01%	
	EF soil	0.00%	
Manufacture of roofing calendered	EF air	0.03%	(European-Chemicals-Bureau 2008)
	EF wastewater	0.03%	
	EF soil	0.00%	
Manufacture of roofing coil coated	EF air	0.09%	(European-Chemicals-Bureau 2008)
	EF wastewater	0.09%	
	EF soil	0.00%	
Manufacture of hoses and profiles	EF air	0.01%	(European-Chemicals-Bureau 2008)
	EF stormwater	0.01%	
	EF soil	0.00%	
Manufacture of car undercoating	EF air	0.11%	(European-Chemicals-Bureau 2008)
	EF wastewater	0.11%	
Manufacture of adhesive and sealant	EF air	0.26%	(European-Chemicals-Bureau 2008)
	EF wastewater	1.00%	
	EF soil	0.51%	
Manufacture of lacquers and paints	EF air	0.25%	(European-Chemicals-Bureau 2008)
	EF wastewater	1.10%	
	EF soil	0.51%	

Manufacture of printing ink	EF air	5.25%	(European-Chemicals-Bureau 2008)
	EF wastewater	1.05%	
	EF soil	0.16%	
Manufacture of ceramic products	EF air	0.25%	(European-Chemicals-Bureau 2008)
	EF wastewater	2.00%	
	EF soil	0.01%	

Supp mat – Table 6: Emission Factors for the use of products

Use of products	Emission Factors expressed in percentages of DEHP contained in products used		References
	Environmental compartment	Value	
Use of flooring	EF air	0.05%	(European-Chemicals-Bureau 2008)
	EF soil	1.10%	
Use of wall covering	EF air	0.05%	(European-Chemicals-Bureau 2008)
Use of coated fabric	EF air	0.05%	(European-Chemicals-Bureau 2008)
Use of film/sheet & coated product	EF air	0.05%	(European-Chemicals-Bureau 2008)
Indoor use of wires and cables	EF air	0.05%	(European-Chemicals-Bureau 2008)
Outdoor use in air use of wires and cables	EF air	0.02%	(European-Chemicals-Bureau 2008)
	EF stormwater	1.57%	
	EF soil	1.57%	
Outdoor in soil use of wires and cables	EF soil	36.00%	(European-Chemicals-Bureau 2008)
Indoor use of moulded product	EF air	0.05%	(European-Chemicals-Bureau 2008)
	EF wastewater	1.00%	
Outdoor use of moulded product	EF stormwater	0.26%	(European-Chemicals-Bureau 2008)
	EF soil	0.26%	
Use of roofing calendered	EF air	0.01%	(European-Chemicals-Bureau 2008)
	EF stormwater	2.44%	
	EF soil	2.44%	
Use of roofing coil coated	EF air	0.05%	(European-Chemicals-Bureau 2008)
	EF stormwater	5.22%	
	EF soil	5.22%	
Indoor use of hoses and profiles	EF air	0.05%	(European-Chemicals-Bureau 2008)
Outdoor use of hoses and profiles	EF stormwater	0.26%	(European-Chemicals-Bureau 2008)
	EF soil	0.26%	
Use of car undercoating	EF air	0.01%	(European-Chemicals-Bureau 2008)
	EF stormwater	0.33%	
	EF wastewater	0.66%	

	EF soil	0.33%	
Indoor use of adhesive and sealant	EF wastewater	1.00%	(European-Chemicals-Bureau 2008)
Outdoor use of adhesive and sealant	EF stormwater	1.00%	(European-Chemicals-Bureau 2008)
Indoor use of lacquers and paints	EF wastewater	14.00%	(European-Chemicals-Bureau 2008)
Outdoor use of lacquers and paints	EF stormwater	7.00%	(European-Chemicals-Bureau 2008)
Use of printing ink	EF air	1.00%	(European-Chemicals-Bureau 2008)
Use of ceramic products	No data		

Supp mat – Table 7: Emissions Factors for the dismantling of products outdoor used

Dismantling of products	Emission Factor expressed in percentages of DEHP contained in the share of products dismantled during all the lifespan		References
	Environmental compartment	Value	
Dismantling of cable during outdoor use	EF stormwater	0.02%	(European-Chemicals-Bureau 2008)
	EF soil	0.05%	
Dismantling of hoses and profiles during outdoor use	EF stormwater	0.05%	(European-Chemicals-Bureau 2008)
	EF soil	0.15%	
Dismantling of coating fabric during outdoor use	EF stormwater	0.10%	(European-Chemicals-Bureau 2008)
	EF soil	0.30%	
Dismantling of moulded products during outdoor use	EF air	0.02%	(European-Chemicals-Bureau 2008)
	EF stormwater	0.50%	
	EF soil	1.50%	
Dismantling of car undercoating during outdoor use	EF air	0.01%	(European-Chemicals-Bureau 2008)
	EF stormwater	0.21%	
	EF soil	0.63%	
Dismantling of roofing calendered and roofing coated products during outdoor use	EF stormwater	0.06%	(European-Chemicals-Bureau 2008)
	EF soil	0.19%	
Dismantling of adhesive and sealant during outdoor use	EF air	0.003%	(European-Chemicals-Bureau 2008)
	EF stormwater	0.063%	
	EF soil	0.188%	
Dismantling of lacquers and paints during outdoor use	EF air	0.01%	(European-Chemicals-Bureau 2008)
	EF stormwater	0.18%	
	EF soil	0.54%	

Supp mat – Table 8: Emissions Factors for end-of-life treatments

End-of-life treatments	Emission Factor expressed in percentages of DEHP contained in the share of products dismantled during all the lifespan		References
	Environmental compartment	Value	
Incineration of products (all types)	EF air	0.00007%	(European-Chemicals-Bureau 2008)
Landfilling of products (all types)	EF soil	0.001884%	Hypothesis based on (COWI 2013) & (Defranceschi., 2015)
Recycling of products (all types)	No data available in the literature		
Shredding of car undercoating	EF air	0.0005%	(European-Chemicals-Bureau 2008)
	EF soil	0.0056%	