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Diverging trends of plasticizers (phthalates and non-phthalates) in indoor and freshwater environments—why?

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Abstract

Background: European chemicals management aims to protect human health and the environment from legacy and emerging contaminants. The plasticizer market changed in response to the restriction of low molecular weight (LMW) phthalate plasticizers such as Di (2-ethylhexyl) phthalate (DEHP) due to their hazardous properties. We investigated patterns and trends of 19 regulated and emerging plasticizers in house dust from German homes and in suspended particulate matter (SPM) from major German rivers. The samples were used from the mid-2000s and late 2010s from two governmental long-term monitoring programs in Germany.

Results: While the sum of the respective plasticizer levels hardly changed over the study period, we observed a significant decrease of LMW phthalates in both house dust (2003/06, 80% of the ∑plasticizer concentration; 2014/17, 31%) and SPM (2005, 48%; 2017, 28%). This was accompanied by their substitution with high molecular weight (HMW) phthalates and non-phthalates. HMW phthalates increased from 19% of the ∑plasticizer concentration to 46% between the mid-2000s and the late 2010s in house dust, and from 50% to 63% in SPM samples. Diisononyl phthalate (DINP) replaced DEHP as the dominant plasticizer in both compartments. A significant tenfold increase ($p < 0.05$) was observed in SPM samples for Di (2-propylheptyl) phthalate (DPHP) (1–13%), compared to low levels in house dust (2014/17, 1%). Non-phthalates increased to 23% of the ∑plasticizer concentration in house dust but only to 9% in SPM (mid-2000s: house dust, < 1%; SPM, 1.5%). In recent house dust samples, Di (2-ethylhexyl) terephthalate (DEHT) had the third highest concentration of all plasticizers and contributed 18% to the total load, whereas Tris (2-ethylhexyl) trimellitate (TOTM) was one of the major non-phthalates in SPM samples.

Conclusions: Unlike in the indoor environment, the substitution of LMW phthalates in the aquatic environment was characterized by a significant shift towards plasticizers with potentially hazardous properties. DPHP and TOTM were identified by European chemical regulation as potentially endocrine disrupting compounds and persistent, bioaccumulative and toxic compounds. Our data document the need for integrated chemicals management to safeguard the transition to a non-toxic environment.

Keywords: EU chemicals policy, Plasticizer, House dust, Environmental monitoring, Integrated exposure assessment

Background

European chemicals policy aims to protect human health and the environment from substances of concern. Precautionary substance and product regulations are in place to prevent the use and marketing of these substances. With the Green Deal [40], the EU is also pursuing new, ambitious goals, such as the zero-pollution action plan for a non-toxic environment. Integral part of

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the zero-pollution action plan is the chemical strategy for sustainability (CSS), which aims to better protect humans and the environment from hazardous chemicals [41]. High-volume industrial chemicals include plasticizers (global production in 2018: 7.45 Mt [20]), the main additives for flexible PVC. The use of some phthalate plasticizers has been successively restricted in the EU since the late 1990s due to their endocrine disrupting (ED) properties, especially in sensitive applications for humans, such as baby products, children's toys, cosmetics and food packaging [31, 33–35]. Di (2-ethylhexyl) phthalate (DEHP), Benzylbutyl phthalate (BBP), Diisobutyl phthalate (DIBP) and Di-n-butyl phthalate (DnBP) are included in REACH Annex XVII, restricted substances list [51], and their use is subject to authorization in the EU since 2015 ("sunset date", [52]). The substitution of the formerly dominant plasticizer DEHP (EU market share 1996: 51%; 2015: 10% [36, 58] has led to a fundamental shift in the market since the 1990s toward HMW phthalates [45], with a simultaneous steady growth in demand for soft PVC [59, 74].

The broad use of plasticizers in large amounts in consumer and industrial products is associated with their occurrence in both the indoor and ambient environment. The ubiquitous presence of DEHP and other low molecular weight (LMW) phthalates such as BBP, DIBP, D*n*BP is well-documented [67, 81, 96, 98, 124]. High molecular weight (HMW) phthalates such as Diisononyl phthalate (DINP) and Diisodecyl phthalate (DIDP) have been detected in indoor air, house dust, blood plasma or in the aquatic environment, including biota [1, 10, 106, 119, 130]. Recent studies confirm the upcoming relevance of non-phthalates in indoor and ambient environment [22, 78, 84, 85, 95, 118].

This work aims to provide: (1) a comparison of patterns and trends of regulated and emerging plasticizers over more than a decade in indoor and freshwater systems and (2) an investigation of the possible reasons for different exposures in the two compartments. Finally, (3) the efficiency and coherence of current EU chemicals regulations—also in view of the new zero-pollution action plan [42].

For this purpose, representative data sets from indoor and freshwater environments were selected with the best match regarding time period, study area and the plasticizer analyte spectrum. The data on plasticizers in house dust originate from the German Environmental Surveys (GerES) IV and GerES V, which were conducted by the German Environment Agency in German households from 2003 to 2006 and from 2014 to 2017. House dust is a sink and repository for semi-volatile organic compounds and it has been used in numerous research programs as a marker of indoor exposure to plasticizers. House dust data are indicative of plasticizer abundance and composition in private households, which are directly influenced by construction materials, personal and lifestyle-products that are used in the indoor environment [13, 114]. For plasticizer exposure in large German rivers, data from measurements in suspended particulate matter (SPM) from the German Environmental Specimen Bank from 2005 to 2017 (provided by the German Environment Agency) were used. SPM is a sink and transport mechanism for solid particles and associated pollutants [77, 97]. Analysis of hydrophobic and lipophilic substances in SPM is an appropriate surrogate for sediment monitoring [37, 108]. Monitoring with SPM is applied in European long-term monitoring programs for EU priority substances [104], including the plasticizer DEHP [76].

To the best of our knowledge, trend data for Di (2-ethylhexyl) terephthalate (DEHT) in European freshwater systems are presented here for the first time.

Methods

Plasticizer selection: legacy phthalates and substitutes of DEHP

At present, about 50 substances with plasticizing properties are in commercial use [60]. Used in combination or individually, plasticizers improve flexibility, extensibility and processability of polymer plastics [23]. Some plasticizers are characterized by weatherability, low-temperature flexibility or heat resistance [68]. Plastizicers are used in a wide range of indoor and outdoor applications including roofing membranes, automotive interiors, electrical cables, cling films, sealants, flooring, wall coverings, food packaging and consumer products [60]. Plasticizers are usually grouped into phthalates and non-phthalates. Phthalates still represent the majority in the European plasticizer market, at around 60% [58]. However, since the end of the 1990s there has been a steady shift within the phthalate plasticizers from LMW phthalates toward HMW phthalates [45]. LMW phthalates are characterized by up to six carbon atoms in the main chain [70]. Furthermore, the demand for alternative plasticizers is increasing. Today, the most frequently used non-phthalates are adipates, terephthalates (main representative DEHT) and Diisononyl 1, 2-cyclohexanedicarboxylic acid (DINCH) [60].

The monitoring programs used in this study covered a total of 26 analytes. For the comparison, only those plasticizers were considered that have been detected in at least two data sets, i.e., 19 plasticizers (12 phthalates and seven non-phthalates). They are listed in Table 1 with their abbreviations, molecular weights and CAS numbers.

Certain highly isomeric phthalates, such as DINP and DIDP, might commercially be available under different

Table 1 Overview on the plasticizers included in this work

Compound	Abbreviation	CAS no	Molecular weight
Phthalates			
Low molecular weight phthalates			
Dimethyl phthalate	DMP	131-11-3	194.2
Diethyl phthalate	DEP	84-66-2	222.3
Di-*n*-butyl phthalate	DnBP	84-74-2	278.3
Diisobutyl phthalate	DIBP	84-69-5	278.3
Benzylbutyl phthalate	BBP	85-68-7	312.4
Dicyclohexyl phthalate	DcHP	84-61-7	330.4
Diisoheptyl phthalate	DIHP	71888-89-6	362.5
Di (2-ethylhexyl) phthalate	DEHP	117-81-7	390.6
High molecular weight phthalates			
Diisononyl phthalate	DINP	28553-12-0, 68515-48-0	418.6
Diisodecyl phthalate	DIDP	26761-40-0, 68515-49-1	446.7
Di (2-propylheptyl) phthalate	DPHP	53306-54-0	446.7
Diisoundecyl phthalate	DIUP	85507-79-5	474.7
Non-phthalates			
Di (2-ethylhexyl) adipate	DEHA	103-23-1	370.6
Diisononyl adipate	DINA	33703-08-1	398.6
Di (2-ethylhexyl) terephthalate	DEHT	6422-86-2	390.6
Acetyl tributyl citrate	ATBC	77-90-7	402.5
Di (2-ethylhexyl) azelate	DEHAz	103-24-2	412.7
Diisononyl 1,2-cyclohexanedicarboxylic acid	DINCH	166412-78-8	424.6
Tris (2-ethylhexyl) trimellitate	TOTM	3319-31-1	546.8

CAS numbers. Additional information on the substances such as SMILES, molecular formula and physicochemical properties are reported in the Supplementary Information (Additional file 1: Table S1). Our comparison excludes Diallyl phthalate (DAP), Di (2-methoxyethyl) phthalate (DMEP), Bis (2-butoxyethyl) phthalate (DBEP), Dipentyl phthalate (DPP), Dihexyl phthalate (DHP), Dibutyl adipate (DBA), Di (2-ethylhexyl) sebacate (DEHS), because they were observed at very low concentrations or below the limit of quantification (LOQ) in one of the studies.

Sampling programs

We compared plasticizer concentrations in Germany from two different, government-funded sampling programs. The first program, the German Environmental Survey (GerES) is a representative study of environmental exposures on the general population of Germany [111], which has been conducted since 1985. GerES investigates, among many other parameters, house dust samples from various indoor environments [110]. The second program, the German Environmental Specimen Bank (ESB), collects and investigates SPM samples from 13 riverine sites in Germany since 2005 (Additional file 1: Fig. S1). Samples are archived continuously in the German Environmental Specimen Bank [62]. The sampling sites reflect different intensities of anthropogenic impacts. Detailed characteristics of both measurement programs are given in Additional file 1: Table S2.

An important aspect of comparing exposure data from indoor and aquatic environments is similar partitioning behavior of plasticizers between SPM and water phase as well as dust and air. Adsorption of phthalate plasticizers to solids increases with their molecular weight and their octanol–water partition coefficient (K_{OW}) [24, 25]. Likewise, the octanol–air partition coefficient (K_{OA}) of phthalates is a strong predictor of their abundance in indoor sinks, such as settled house dust relative to their airborne concentration [125, 126] (Additional file 1: Table S1) for K_{OA} and K_{OW} of the plasticizers investigated). The fate of plasticizers can differ in indoor and aquatic environments, because environmental conditions are not comparable. The predominant fate of phthalates in aerobic aquatic environments is biodegradation [102], whereas losses in indoor environments caused by microbial degradation could only be demonstrated at elevated humidity [16]. However, it can be expected that these fundamental processes, e.g., sorption, degradation half-life, do not

change with time. Even if a plasticizer adsorbs to a greater extent to house dust than to SPM, this will be meaningful to the plasticizer load in the respective compartment, but not for its temporal trend. Since the sampling methods for house dust and suspended particulate matter are quality assured and highly conserved [63, 121], the data of the two study programs can accurately reflect temporal trends of the plasticizers.

Plasticizers in house dust

In each of the GerES study cycles (GerES IV: 2003–2006; GerES V: 2014–2017), house dust samples were collected in a number of about 600 households spread across 150 study locations across Germany (Additional file 1: Fig. S1). The data from GerES IV (2003–2006) have been previously reported [92–94]. The data from GerES V (2014–2017) have been unpublished, but were generated using the same procedures.

In both cases, the sampling strategy followed a random selection procedure, which provided for population-representative house dust samples [110]. In GerES IV the mass fractions of 12 phthalates and 9 non-phthalates were determined. Briefly, the content of each vacuum cleaner bag was sieved to obtain the fraction of particles < 63 µm. This fraction was chosen to reduce the variability of the analytical results [91]. Chemical analysis of semi-volatile organic compounds (SVOC) in this fraction in GerES IV were performed by [92–94]. Plasticizer concentrations were determined using liquid chromatography/mass spectrometry (LC–MS) with LOQs between 0.08 and 16.5 µg/g (Additional file 1: Table S7a). For GerES V, corresponding work was performed by the Fraunhofer Institute for Process Engineering and Packaging IVV in Freising, Germany. Extracts of the GerES V data have been reported in conjunction with GerES V human biomonitoring data in Schwedler et al. [112].

Plasticizers in riverine suspended particulate matter

To compare the long-term pattern of plasticizers in indoor and freshwater systems, existing SPM data from a study on spatial and temporal trends of plasticizers in German rivers were used [95]. In the study, archived SPM samples from the ESB have been analyzed for 23 plasticizers (17 phthalates and 6 non-phthalates). The samples were collected between 2005 and 2017 at sampling sites in major German rivers, such as the Rhine, Elbe, Danube and tributaries, i.e., Saar, Saale and Mulde. A brief characterization of the sampling sites is given in Additional file 1: Table S3. Suspended particles are collected in sedimentation boxes and sampled on a monthly basis. 12 monthly samples are than pooled to one annual sample. The samples are sieved (< 2 mm), homogenized and freeze-dried (more details in the Additional file 1; [63]).

For the present work, data for the 19 plasticizers (Table 1) investigated at 11 sites in 2005 (Saar sites in 2006) and 2017 were used. The Danube sites are not included, because sampling started only in 2009.

An LC-based method has been applied for the analysis of plasticizers, which did not cover DEHT as one of the potential DEHP-substitutes. The determination of DEHT in SPM samples was performed separately by gas chromatography/mass spectrometry (GC–MS) analysis. Sampling strategy and sampling areas are identical to the study of Nagorka and Koschorreck [95]. The analytical method was based on a solvent extraction of the SPM samples. After clean-up with an alumina column the eluates were measured by gas chromatography/mass spectrometry. Details on chemicals and materials are given in the Additional file 1. In addition, parameters for extraction, clean-up, GC–MS analysis and the quality assurance can be found in the Additional file 1.

Statistics

Mean and median values of the same 19 plasticizers from the house dust and SPM studies were used for the comparative description. For harmonization, values below the LOQ were considered with ½ LOQ, although a different approach for values below the LOQ from GerES IV samples had been used in the original papers (replacement by ⅔ LOQ; [92–94]).

For house dust samples, the significance of the temporal mean differences was calculated using the Mann–Whitney *U* test (95% confidence interval, significance level $p < 0.05$). In the indoor data sets, the LOQs for the individual analytes differed both within GerES V and between GerES IV and GerES V. Prior to testing for statistical significance, a constant LOQ was established for each plasticizer. The highest LOQ (LOQ_{max}) of the respective analyte was determined for all indoor data. All measured values below LOQ_{max} were replaced by ½ LOQ_{max}. For individual analytes, more than 50% of the measured values were below the LOQ in both study cycles, so that a calculation of the Mann–Whitney *U* statistic was not meaningful. The SPM sampling sites were identical in 2005 and 2017; therefore, a paired test was applied for SPM data. Due to the small sample size, the means were compared by a non-parametric test (Wilcoxon signed-rank test, significance level $p < 0.05$). No significance tests were applied for plasticizers analyzed with a detection frequency of less than 50%.

Results and discussion

House dust and freshwater environment—a comparison of plasticizer levels and pattern

LMW phthalates

We noticed a decrease of LMW phthalates (including DEHP) in samples from house dust (2003/06, 80% of the ∑plasticizer concentration; 2014/17, 31%) and

SPM (2005, 48%; 2017, 28%). The significant reduction ($p < 0.05$) of DEHP levels was accompanied by its substitution with HMW phthalates and non-phthalates, resulting in a change in the pattern of substances in indoor and aquatic environments (Figs. 1a, b and 2). Overall, the sum of plasticizer levels hardly differed over the study period in both matrices: In house dust, the temporal change was significant, but small (mean from 1194 µg/g to 1208 µg/g, median from 981 µg/g to 855 µg/g; $p < 0.05$). No significant differences were found in SPM samples between 2005 and 2017 (mean from 6160 ng/g dry weight (dw) to 4400 ng/g dw; median from 3600 ng/g dw to 33220 ng/g dw; $p > 0.05$). Consequently, the decline of LMW phthalates does on average not noticeable reduce the total plasticizer load in both compartments. However, in detail we found diverging developments in house dust and SPM. Descriptive statistics on the plasticizers included in this comparative analysis are given in Additional file 1: Tables S7a, b. The test results for statistical significance comparing the mean difference between concentrations in the mid-2000s and in the mid-2010s are presented in Additional file 1: Table S8.

In samples from the mid-2000s, DEHP was the overall dominant compound in both matrices (Fig. 1a, b) and LMW phthalates accounted for 80% of the ∑plasticizer content in house dust (DEHP, 57%; C1–C7-phthalates, 23%) and 48% in SPM samples (DEHP 44%; C1–C7-phthalates 4%), respectively. In the samples from the mid-2010s, the percentage of LMW phthalates in both indoor and aquatic samples was much more similar (house dust, 31%; SPM, 28%). Furthermore, the DEHP percentages in both compartments neared a comparable level (house dust, 21%; SPM, 23%). Mean and median DEHP values have dropped by more than half compared to the mid-2000s ($p < 0.05$) (house dust, mean from 685 µg/g to 250 µg/g; SPM, mean from 2710 ng/g dw to 991 ng/g dw). In house dust, C1–C7-phthalates followed this trend and their levels halved since the mid-2000s (mean from 271 µg/g to 121 µg/g), due to significantly decreasing levels of DEP, DBP (∑concentration of DnBP and DIBP), BBP and DIHP. This corresponds to a decline of the median values by two thirds compared to the values from 2003/06. In contrast, in SPM, the concentration of C1–C7-phthalates remained almost unchanged (mean from 249 µg/g to 244 µg/g; median from 250 ng/g dw to 263 ng/g dw) as a decrease ($p < 0.05$) in BBP levels was offset by increasing DBP levels, which were, however, not statistically significant. In response to regulatory classifications as toxic, the European market shares have already decreased between 1998 and 2008 (BBP, from 3% to 1%; DBP, from 6% to 3%) [122]. BBP was widely used in the flooring industry [88], whereas primary uses of DnPB included indoor and outdoor applications, such as consumer goods, adhesives, lacquers, paints and sealants [8]. Because DIBP has similar properties as DnBP, it could be used as a substitute [29]. DBP contents up to 60% were found in foams used in grouting applications for water control in tunnels and sewer systems [88]. In addition

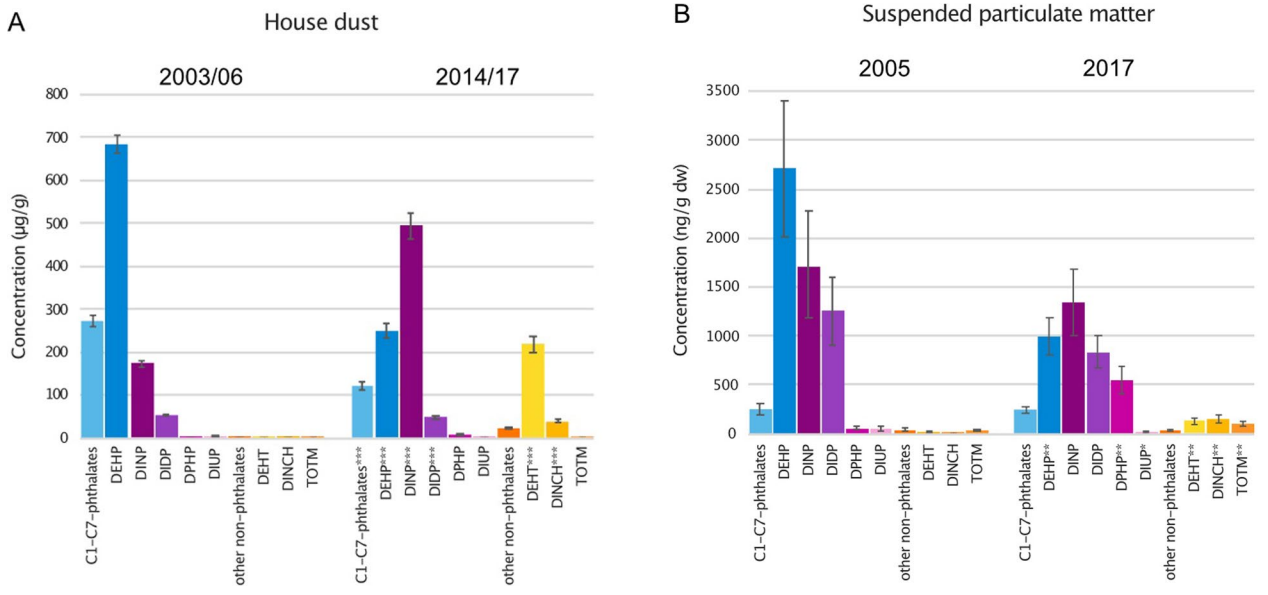

Fig. 1 **a**, **b** Plasticizer mean concentrations in house dust and SPM samples in mid-2000s and in 2017; bars with standard error; plasticizer with marking, significance level, *$p < 0.05$, **$p < 0.01$, ***$p < 0.001$; without marking, $p > 0.05$

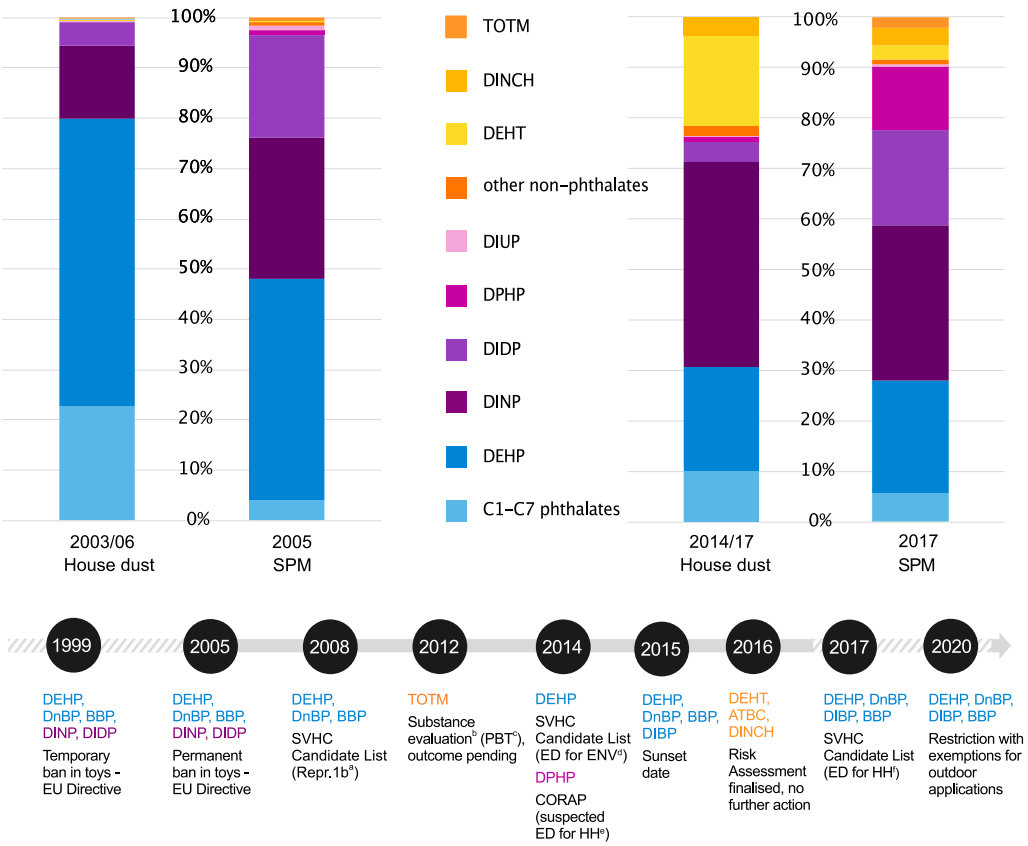

Fig. 2 Relative compositions of plasticizers in house dust and riverine SPM samples in the mid-2000s and in 2017; [a]toxic for reproduction; [b]substance evaluation under REACH; [c]persistent, bioaccumulative, toxic; [d]endocrine disrupting (ED) properties with effects on the environment; [e]suspected ED properties with effects on human health; [f]ED properties with effects on human health. *SVHC* Substance of Very High Concern, *CoRAP* Community Rolling Action Plan

to direct and indirect release into the environment from product uses, atmospheric deposition is suggested to be a relevant exposure pathway for DBP [128, 129].

The data indicate continuous substitution of critical phthalates such as DEHP, DBP and BBP in response to increasing regulatory action on plasticizers in Europe, which began in 1999 (Fig. 2) and concluded with the need to apply for authorization for the continued use of those plasticizers in 2015, after the so called "sunset date". This is in good agreement with human biomonitoring studies in Germany, where urine samples show a significant decrease since 1999/2000 [81, 112]. The reduction may be attributed to the displacement of DEHP and very LMW phthalates from exposure sources, such as consumer products and food contact products. A similar rapidly declining trend for LMW phthalates was observed in household dust in Taiwan between 2006 and 2014 as a result of chemical management there [75].

HMW phthalates

Over the study period, the percentage of HMW phthalates in house dust increased from 19% of the ∑plasticizer concentration to 46% and in SPM samples from 50% to 63%. DINP became the main plasticizer in both compartments. In dust samples, DINP levels increased significantly over the study period (mean from 173 µg/g to 494 µg/g). By contrast, a significant decrease was observed for DIDP (mean from 54 µg/g to 48 µg/g) and the concentrations of DPHP and DIUP remained low (DPHP, 1% of the ∑plasticizer concentration; DIUP, 1%). In samples from mid-2010s, the concentrations of DINP were 10 times higher than those of DIDP. The predominant presence of DINP in dust samples from 2014/17 may result from its increasing use in vinyl flooring, wall covering and furniture [32, 44]. Studies in indoor

environments have demonstrated that PVC flooring and wallpapers are associated with higher DINP levels in dust [2, 22, 84].

In SPM, DINP concentrations and patterns hardly changed over the study period (mean from 1730 ng/g dw to 1340 ng/g dw; share from 28% to 31%). Nevertheless, it became the main compound in the SPM samples as the DEHP content continuously decreased. In the mid-2000s, DIDP percentages in SPM samples were four times higher than in house dust, but did not change much since then (2005, 20%; 2017, 19%). Concentrations for DIUP were low (< 1% of the ∑plasticizer concentration) compared to other HMW phthalates, and moreover, a significant decrease was observed. However, we observed a striking tenfold increase ($p < 0.05$) in DPHP concentrations (mean from 57 ng/g dw to 550 ng/g dw) in SPM samples, although DPHP has been under regulatory scrutiny and included on the Community Rolling Action Plan (CoRAP) list since 2014. (Fig. 2; [46, 47]) DPHP was advertised in the early 2000s as an DEHP substitute for outdoor applications [82], and like DIDP, it is used for the manufacture of wires, cables, hoses and tarpaulins [44]. Its weather resistance and low migration rates give DPHP advantages over other DIDP specifications for the use in outdoor applications, such as cladding and roofing [101, 117]. Although DPHP is registered for both outdoor and indoor use in durable materials with low release rates [53], our house dust data suggest that it contributes only little to indoor exposure (2014/17, 1% of the ∑plasticizer concentration).

Non-phthalates

The use of non-phthalates in the European plasticizer market was almost constant between 1999 and 2005, but then increased from 7% of the ∑plasticizer concentration to 40% in 2017 [17, 59]. In the mid-2000s, the share of non-phthalates was comparatively low (house dust, < 1%; SPM, 1.5%). The indoor samples reflected the shift from phthalates to non-phthalates more pronouncedly than the SPM samples. In the mid-2010s, non-phthalates accounted for 23% in house dust, but only for 9% in SPM.

Also, the pattern of non-phthalates in the more recent samples differed significantly in the samples from the two compartments. In house dust samples from mid-2010s, the mean values of DEHT and DINCH have increased by a factor of 90 and 40, respectively, compared to samples from the mid-2000s. DEHT became the third highest fraction (mean 220 µg/g) and contributed 18% of the total load, while DINCH and ATBC stayed much less relevant with 3.5% and 1.5%, respectively. TOTM, an extremely low volatile plasticizer was only detected at low levels in house dust. Trends for ATBC could not be verified, because ATBC was not part of the analyte spectrum in GerES IV, the first house dust sampling cycle. However, indicative data from 2009 resulted in a GM of 2.4 µg/g and an arithmetic mean of 6.5 µg/g [93], which were lower values than the results presented here for 2014/17, suggesting a rise in ATBC levels. In general, non-phthalates, such as DEHT, ATBC, and DINCH, which was introduced in 2002, are preferably used as DEHP substitutes in sensitive, consumer-oriented applications, such as toys for children under the age of 3, soft plastic shoes, medical devices, and pharmaceutical coatings [4, 38, 123].

In contrast to house dust samples, the plasticizer pattern in SPM samples from mid-2010s is still very much dominated by phthalates (2017, 91% of the ∑plasticizer concentration). We identified emerging non-phthalates, TOTM, DEHT and DINCH, each with 2–3.5% of the total content. Although ATBC also showed an increasing trend in these samples [95], the levels remained comparably low in 2017 compared to the other DEHP substitutes. For TOTM, exposure pathways in the environment independent of urban waste water can be assumed, since it had limited relevance in house dust samples. TOTM is typically applied in the automotive industry and in cable insulations [61]. In samples from the river Mulde, TOTM accounted for 0.5% of the total plasticizer burden in 2005 and for 9% in 2017. The sampling site is located downstream the Bitterfeld–Wolfen Chemical Park, one of the largest areas for the chemical industry in Eastern Germany. Increasing TOTM levels can be caused by growing industrial settlement in this area after a restructuring process [9, 21]. In highly industrialized bays in Korea TOTM had a share of 36% of the overall plasticizer concentration in sediments which were characterized by automotive plants [79].

Current studies of house dust in various European indoor environments (households, kindergartens, schools, offices) show comparable results as presented here: For example, Larsson et al. also detected DINP, DEHP, DEHT, and DINCH as major plasticizers in dust from 100 Swedish schools and preschools (2017) [84]. Similarly, DINP, DEHP, DEHT, and DINCH were the dominant components in domestic indoor environments from Central/Northern Europe [22, 30]. The proportion of non-phthalates (up to 45% of ∑plasticizers) was even higher than in the GerES V dust samples. Data from other EU countries suggest, that there may be country-specific differences, e.g., in households in Belgium, DINCH exposure was 21% and in house dust from apartments in Stockholm, ATBC was the major non-phthalate besides DEHT [127]. Comparison with data from outside the EU is challenging. Chemical management varies considerably between regions and on a global scale.

DEHP still dominates the plasticizer market [83]. Therefore, target analytes were often restricted to DEHP and other LMW phthalates. House dust samples in China, for example, were strongly dominated by DEHP and DBP [87]. Studies in Thailand and Quatar revealed a plasticizer pattern with DEHP as the main plasticizer, closely followed by DINP [3, 105]. In contrast, DEHT, ATBC and DEHP were the plasticizers with the highest concentrations in US homes [73, 114, 118]. Bi et al. [12] detected DEHP levels in US low-income homes, which were comparable to our results (median, 155 µg/g and mean, 271 µg/g). Similar to the EU restrictions, U.S. Consumer Product Safety Commission (CPSC) had banned the use of DEHP, DBP and BBP in toys and childcare articles in 2008 [28]. Recent investigations on plasticizer levels in sediments from non-industrial coastal areas in Korea are in good agreement with our data and confirm a low proportion of non-phthalates (3–10%) of the total plasticizer burden in aquatic environments [78, 85]. DINP and DEHP were the main phthalates. Among the non-phthalates, the highest concentrations were found for TOTM and DEHT [80]. DEHT and TOTM were detected at levels similar to our data (DEHT, mean 141 ng/g dw; TOTM, mean 150 ng/g dw). DPHP, the major substitute of DEHP in the German SPM samples, was not analyzed in these studies. Several studies have documented the dominant occurrence of DEHP in the mid-2000s in house dust and in sediments or suspended particulate matter from aquatic environments [11, 64, 100, 107, 113, 124]. However, literature data on HMW phthalates and non-phthalates in indoor and riverine samples from this period are largely missing. Sediment studies in Sweden in 2006 found levels for DEHP, DINP, and DIDP that were comparable to our 2005 SPM data (DEHP, 82–2800 ng/g dw; DINP, 130–3200 ng/g dw; DIDP, 190–3400 ng/g dw) [26]. In this study DEHA was the only non-phthalate analyzed (68–520 ng/g dw).

Regulatory discussion of the data

Regulatory assessment of plasticizers

Figure 3 presents the current regulatory status of plasticizers investigated here in the EU. From the 19 substances that were found abundant in samples from the German indoor and aquatic environment, six LMW phthalates (DEHP, DnBP, DIBP, BBP, DcHP, DIHP) have so far been identified as substances of very high concern (SVHC) [54]. Four of those (DEHP, DnBP, DIBP, BBP) have also been restricted under REACH [51]. Two further HMW phthalates, DINP and DIDP, are banned from use in toys, which can be taken into mouth [55]. However, both substances are not identified as SVHC [49, 56]. An overview of the different regulatory processes in the EU for all substances listed in Table 1 can be found in Additional file 1: Table S6a, b and Fig. S2. In the 1980s, first concerns were raised over the health effects of plasticizers and the need for chemical management [66, 120]. The example of DEHP chemical management in Europe shows that it can take almost 20 years to regulate a compound of concern, from the first report of a toxicological concern to the initial provisional EU-wide regulation for phthalates in toys. In 1999, the European Commission (EC) imposed a temporary ban on the use of DEHP together with five other plasticizers (DnBP, BBP, Di-n-octyl phthalate (DnOP), DINP, DIDP) in children's toys that can be put in the mouth [31]. Similar steps have been taken by regulatory bodies in other regions [19, 28, 90]. Only in 2014, DEHP was identified as SVHC because of its ED properties in the environment [48]. Along with DnBP, DIBP and BBP, an identification as SVHC due to ED properties to human health followed in 2017 [39]. For the first time, a substance risk assessment of plasticizers identified a concern about ED effects in both, humans and in the environment. Overall, from 1999 it took another 20 years to establish an—almost—complete ban of DEHP in the EU (Fig. 2, for a more detailed timeline Additional file 1: Fig. S2). The far greater share of the plasticizers (incl. all non-phthalates) has either not been subject to any kind of regulation (i.e., authorization or restriction) or are still in the process of being evaluated by authorities (Fig. 3).

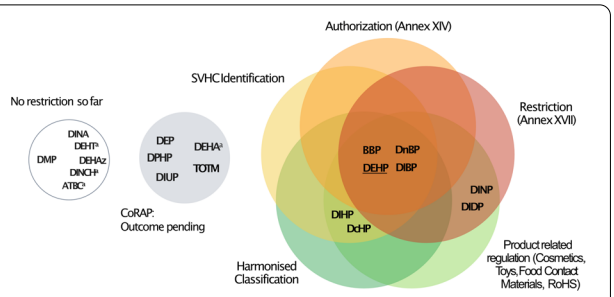

Fig. 3 Present regulations of the plasticizers investigated in this work under the EU chemical management; [a]authorized for Food Contact Materials; *SVHC* Substance of Very High Concern, *CoRAP* Community Rolling Action Plan, *RoHS* Restriction of (the use of certain) Hazardous Substances in electrical and electronic Equipment

In 2012, TOTM had been the first non-phthalate plasticizer for which a substance evaluation had been initiated due to its suspected persistent, bioaccumulating and toxic (PBT) properties (Fig. 2; [43]). Furthermore, a potential ED concern for the environment was identified in 2020 [50]. Substances that persist for long periods of time in the environment and have a high potential to accumulate in biota are of specific concern, because their long-term effects are rarely predictable [57]. Once released into the environment, exposure to these substances is very difficult to reverse, even if emissions are

Fig. 4 **a**–**d** Scatterplots of molecular weight and hydrophobicity (log K_{OW}) showing the relative differences and trends between house dust and SPM samples (DMP, DINA and DEHAz are not pictured due to their constantly low concentrations); circular areas are normalized to 100%; log K_{OW} values were determined with the quantitative structure–activity relationship (QSAR) model JANUS (Additional file 1: Table S1; [103])

stopped. The prevention of irreversible exposure is a central element of the precautionary principle in the European chemicals regulation and the backbone of existing PBT/vPvB regulation [27]. Our data show increasing TOTM concentrations in both indoor and freshwater environments and warrant rapid finalization of the regulatory PBT and ED assessment, which has now been ongoing for nearly a decade. Similarly, in 2014, DPHP was included in the CoRAP [46, 47] due its suspected ED effects on human health. It was not until six years later that the substance evaluation for DPHP was initiated under REACH. During this period, both use [109] and the exposure of the aquatic environment to DPHP have increased manifold. It is important to note that the identification as an SVHC and inclusion in the SVHC Candidate List does not directly correspond to a ban or a strict regulation of the substance.

In 2016, the outcome of an assessment of the non-phthalates DEHT, ATBC and DINCH was published by the evaluating member state authority concluding that no further regulatory action was deemed necessary at the moment [5–7]. The initial reason of concern for the assessment was that those non-phthalates are known, or suspected, to be used as an alternative to phthalates that were previously identified as SVHC and included in the Candidate List (e.g., DEHP). Independent of the outcome of the REACH assessment, our data show the ubiquitous presence of these compounds especially in indoor environments and, to a lesser extent—in freshwater environments.

Molecular weight shift

In samples from both compartments, we observed a shift towards plasticizers with higher molecular weight during the study period (Fig. 4a–d). In house dust samples,

the place of LMW phthalates has been taken in particular by DINP as well as DEHT and DINCH, which have at least the same molecular weight as DEHP (DEHT) or a slightly higher one (DINP, DINCH). In samples from 2014/17, these three substances account for 62% of the total load. Their use is not subject to any restrictions with a few exceptions (DINP; Additional file 1: Table S6a, b). We did not observe a replacement of LMW phthalates by other short-chain non-phthalates, e.g., DBA (see "Methods" chapter 'Plasticizer selection: legacy phthalates and substitutes of DEHP'), which were also considered as potential substitutes for regulated phthalates [14]. Other European studies support this finding [22, 65]. In SPM samples, the growth rate of higher molecular weight phthalates and non-phthalates is less prominent than in house dust samples. However, HMW phthalates already constituted a comparatively strong proportion in the mid-2000s. In general, plasticizers with low volatility and migration rate are more suitable for stressed outdoor applications with respect to hydrolysis, leaching and microbial degradation [72, 116]. However, the increase of DPHP and TOTM during the study period was partially compensated by slightly decreasing DINP and DIDP contents. In contrast to the indoor environment, the substitution of DEHP in freshwater environments is accompanied by a noticeable shift towards potentially critical plasticizers. The percentage of plasticizers classified as a potential concern in the investigated riverine systems is now 16%, compared to only 2% in house dust. The proportion of plasticizers in SPM samples, which are not classified as hazardous, accounted for 56% compared to 68% in house dust samples.

We suspect that this trend will be further continued in freshwater environments, since DPHP in particular is increasing rapidly in the SPM samples. HMW phthalates are usually marketed as moderate or highly branched isomer mixtures. As reported, the water solubility of phthalates with branched chains is higher than of those with linear chains [71, 86]. Therefore, leaching can be expected even for very low volatile plasticizers which is supported by our data. In a long-term study of geomembranes installed on Italian dams, plasticizer losses of up to 22% were observed [18]. Substitutes such as DEHT, DINCH, and ATBC are less recommended for outdoor building and construction applications due to their poor weathering properties and high migration rate (ATBC) [88, 99, 115]. In addition, terephthalate esters (e.g., DEHT) in PVC are less compatible and more difficult to process than conventional phthalates with the same carbon atom number [69].

Conclusions

The data show that over the past 20 years, the chemical industry has modified their production and marketing strategies by substituting the former "blockbuster" plasticizer DEHP in indoor and outdoor applications. Our findings suggest that non-toxic chemicals, such as DEHT, are becoming more abundant in indoor materials and products, while low volatile, potentially hazardous plasticizers such as DPHP and TOTM with suspected PBT and ED properties seem to have become the new additives for PVC in outdoor applications. We hypothesize that EU chemical management for plasticizers has put a focus on human health related concerns. Indeed, product-specific regulations between 1999 and 2007 were exclusively related to consumer goods and food packaging materials. In 2007, REACH entered into force and up to now, REACH has initiated 27 regulatory processes for the plasticizers investigated in this work (group entries were counted as one process). Only six of these activities address environmental impacts and two EU regulations focusing on environmental concerns have been introduced up to date (Water Framework Directive (WFD), Waste Framework Directive). Furthermore, some REACH regulations include exemptions for certain uses of plasticizers, where no consumer exposure is expected but environmental exposure is likely to occur (e.g., the wide-scope restriction of DEHP, DnBP, DIBP and BBP exempts articles exclusively for industrial or agricultural use or use in open air) [51].

Domestic product use as a source for plasticizer contamination in surface waters cannot be excluded. However, other studies support the assumption that continuous discharge of urban wastewater from residential areas is not the main contributing factor [15]. Malnes et al. reported comparable levels of ATBC in wastewater-impacted Swedish rivers and in a lake with no wastewater discharge (2022) [89]. The highest plasticizer concentrations, and likewise, the highest TOTM levels in sediments from Korean bays were found at sampling sites, which were surrounded by automobile, construction and petrochemical industry [79].

Due to its ubiquity in the environment, plasticizers have raised concerns pertaining to continuous exposure of the human population. Our data/observations document the need for an integrated exposure assessment in the sense of the precautionary principle in chemicals management, as well as toxicity assessment/identification of hazard arising from the new introduced substances. In the long term, a stronger focus on regulation, which is largely based on human health concerns will turn out to be detrimental for both protection goals due to the fact that humans also can be exposed to hazardous substances via the environment. However, up to now this

overlap remains largely unrecognized in the different chemical regulations and environmental risks are not sufficiently addressed in risk assessment.

Abbreviations

ATBC: Acetyl tributyl citrate; BBP: Benzylbutyl phthalate; CAS: Chemical Abstracts Service; CSS: Chemicals Strategy for Sustainability; CoRAP: Community Rolling Action Plan; DAP: Diallyl phthalate; DBA: Dibutyl adipate; DBEP: Bis (2-butoxyethyl) phthalate; DEHA: Di (2-ethylhexyl) adipate; DEHAz: Di (2-ethylhexyl) azelate; DEHP: Di (2-ethylhexyl) phthalate; DEHS: Di (2-ethylhexyl) sebacate; DEHT: Di (2-ethylhexyl) terephthalate; DEP: Diethyl phthalate; DBP: Dibutyl phthalate; DcHP: Dicyclohexyl phthalate; DIBP: Diisobutyl phthalate; DIDP: Diisodecyl phthalate; DHP: Dihexyl phthalate; DIHP: Diisoheptyl phthalate; DINA: Diisononyl adipate; DINCH: Diisononyl 1, 2-cyclohexanedicarboxylic acid; DINP: Diisononyl phthalate; DIUP: Diisoundecyl phthalate; DMEP: Di (2-methoxyethyl) phthalate; DMP: Dimethyl phthalate; DnBP: Di-*n*-butyl phthalate; DPHP: Di (2-propylheptyl) phthalate; DPP: Dipentyl phthalate; dw: dry weight; ED: Endocrine disruptor; ESB: German Environmental Specimen Bank; EU: European Union; GerES: German Environmental Survey; GC–MS: Gas chromatography/mass spectrometry; HMW: High molecular weight; LC–MS: Liquid chromatography/mass spectrometry; LOQ: Limit of quantification; K_{OA}: Octanol–air partition coefficient; K_{OW}: Octanol–water partition coefficient; LMW: Low molecular weight; PBT: Persistent, bioaccumulative, toxic; PVC: Polyvinyl chloride; QSAR: Quantitative structure–activity relationship; REACH: Registration Evaluation Authorisation and Restriction of Chemicals; RoHS: Restriction of (the use of certain) Hazardous Substances in electrical and electronical Equipment; SMILES: Simplified Molecular Input Line Entry Specification; SPM: Suspended particulate matter; SVHC: Substance of Very High Concern; SVOC: Semi-volatile organic compound; TOTM: Tris(2-ethylhexyl) trimellitate; WFD: Water Framework Directive.

Supplementary Information

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Additional file 1: Table S1. General information on the plasticizers investigated. **Table S2.** Overview of the sampling programs used for this work. **Figure S1.** Study locations for the German Environmental Survey (GerES) and SPM sampling points for the German Environmental Specimen Bank (ESB). **Table S3.** Sites of the ESB for SPM sampling. **Table S4.** Information on GC/MS system and parameters. **Table S5.** Recoveries for DEHT at different concentration levels. **Table S6a.** Specific product regulations. **Table S6b.** REACH regulation. **Figure S2.** Relevant regulations for the plasticizers investigated since 1999. **Table S7a.** Characteristic values of the plasticizer investigated in house dust samples. **Table S7b.** Characteristic values of the plasticizer investigated in SPM samples. **Table S8.** Summary of test data and significance of the differences.

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Author contributions

RN, JK and JS have closely collaborated to write the first draft manuscript. WB provided detailed feedback/material on specific chapters. All authors have made comments. All authors read and approved the final manuscript.

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Availability of data and materials

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Declarations

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Consent for publication

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Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence this work reported in this paper.

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