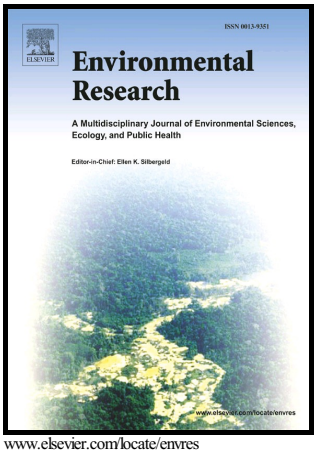
Occurrence of legacy and alternative plasticizers in indoor dust from various EU countries and implications for human exposure via dust ingestion and dermal absorption



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Occurrence of legacy and alternative plasticizers in indoor dust from various EU countries and implications for human exposure via dust ingestion and dermal absorption

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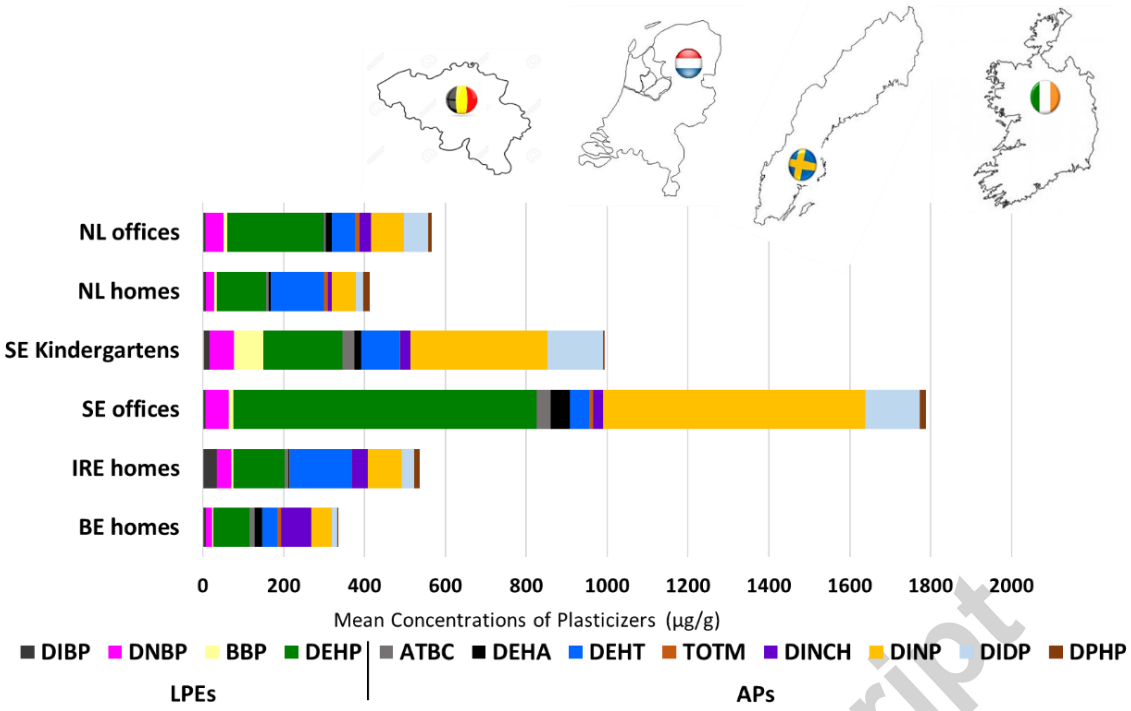
Graphical abstract



Abstract

Plasticizers are a category of chemicals extensively used in consumer products and, consequently, their presence is ubiquitous in the indoor environment. In the present study, an analytical method has been developed for the quantification of plasticizers (7 legacy phthalate esters (LPEs) and 14 alternative plasticizers (APs) in indoor floor dust based on ultrasonic and vortex extraction, Florisil fractionation and GC-(EI)-MS analysis. Dust samples (n=54) were collected from homes, offices, and daycare centers from different EU countries (Belgium, the Netherlands, Ireland and Sweden). Method LOQs ranged from 0.2 to 5 µg/g. Tri-n-hexyl trimellitate (THTM) was not detected in any sample, whereas dimethyl phthalate (DMP), diphenyl phthalate and acetyl triethyl citrate (ATEC) were detected only in 6, 2 and 1 out of 54 samples, respectively. The highest concentrations of plasticizers were measured in Swedish offices, at a mean concentration of total plasticizers of 1,800 µg/g, followed by Swedish daycare centers at 1,200 and 670 µg/g for winter and spring sampling, respectively. Generally, the contribution of APs was slightly higher than for LPEs for all indoor environments (mean contribution 60 and 40%, respectively based on contributions per indoor environment). For the APs, main contributors were DINP in Belgian homes (28%), Swedish offices (60 %), Swedish daycare centers (48%), and Dutch offices (31 %) and DEHT in Belgian (28%), Irish (40%) and Dutch homes (37%) of total APs. The predominant LPE was bis-2-ethylhexyl-phthalate (DEHP) with a mean contribution varying from 60 to 85% of total LPEs. Human exposure was evaluated for

dust ingestion and dermal absorption using hazard quotients (HQs) of plasticizers (ratio between average daily doses and the reference dose). None of the HQs of plasticizers exceeded 1, meaning that the risk for adverse human health effects from these plasticizers via dust ingestion and dermal absorption is unlikely.

Keywords: phthalates, plasticizers, indoor dust, human exposure

1. Introduction

Plasticizers are a major category of chemical additives incorporated into polymers to facilitate their ease of processing and provide specific characteristics to the final product, such as durability, elasticity and flexibility (Bergh et al., 2011). Phthalic acid esters, or phthalates, are major plasticizing agents added mainly to polyvinyl chloride (PVC) material and, to a lesser extent, to non-PVC products. In 2012, more than 90% of all phthalates in Europe were used in the production of PVC (KEMI, 2015). The main representative compounds of this group are dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (DNBP), di-iso-butyl phthalate (DIBP), butyl-benzyl-phthalate (BBzP), di-isononyl phthalate (DINP), di-isodecyl phthalate (DIDP), bis-2-ethylhexyl phthalate (DEHP) and bis (2-propylheptyl) phthalate (DPHP).

Phthalates are semi volatile organic compounds (SVOCs) and they do not form chemical bonds with the polymeric material, so they tend to migrate from the products to the indoor environment, and especially into dust, due to their lipophilic characteristics (Subedi et al., 2017). That means that they can enter easily human body via accidental dust ingestion, inhalation or dermal absorption and further metabolized (Giovanoulis et al., 2017). Human exposure to plasticizers is currently a hot topic for environmental and human health. In the case of phthalates, the toxicity is basically attributed to the metabolites formed, which are more toxic than the parent compounds (Eljezi et al., 2017; Xiang et al., 2017; Barber et al., 1994).

According to literature studies, phthalates have shown adverse effects on human health, for example, effects on reproduction, endocrine disruption (Rudel et al., 2003; Liu et al., 2012) and links to increased incidence of allergies and asthma in children (Braun et al., 2013). They are currently included in Annex XVII, restricted substances list, of REACH (REACH, 2018). DEHP, DNBP, BBzP, DINP and DIDP in PVC products, toys and childcare articles must not exceed 0.1% of the plasticized material (REACH, 2018; ECHA, 2010). In addition, DEHP has been listed in category 1B of substances known, or presumed, to have carcinogenic effects in animals, based on the Globally Harmonized System of classification and labelling of chemicals (GHS) (Bui et al., 2016). Phthalates are also restricted in Canada and U.S.A., with regulatory limits on the application of DEHP, DINP and DIDP in children's products (Snijder et al., 2012; USCPSC, 2007).

These limitations have led inevitably to an increasing need for alternative compounds which meet the market standards but also have lower migration rates and toxicity. Now, current-use or alternative

plasticizers (APs) include several classes: e.g. adipates, terephthalates, trimellitates, citrates, sebacates, cyclohexane dicarboxylic acid esters and phosphates (Supplementary Material, Fig. S1). The main APs in use are di-isobutyl adipate (DIBA), acetyl triethyl citrate (ATEC), di-isobutyl sebacate (DBS), tributyl-O-acetyl citrate (ATBC), bis-2-ethylhexyl-adipate (DEHA), diphenyl cresyl phosphate (DPCP), n-butyryl-tri-n-hexyl citrate (BTHC), bis-2-ethylhexyl-terephthalate (DEHT), tri-n-hexyl trimellitate (THTM), tris (2-ethylhexyl) trimellitate (TOTM), and di-iso-nonyl ester 1,2-cyclohexane dicarboxylic acid (DINCH). Toxicological information concerning phthalates is largely available, but for the APs, there is still a lack of information (EU, 2011).

The present study is a part of the CEFIC project SHINE (Target and Non- Target screening of chemicals in the indoor environment for human exposure assessment) (CEFIC, 2016) in order to provide information of targeted analysis of emerging contaminants in dust collected in schools/daycare centers, homes and offices in various European countries. Two groups of plasticizers were considered: legacy phthalate esters (LPEs) concerning 7 target compounds (DMP, DEP, DNBP, DIBP, BBzP, DEHP, DPP) and alternative plasticizers (APs) concerning 14 target compounds (DIBA, ATEC, DBS, ATBC, DEHA, DPCP, BTHC, DEHT, THTM, TOTM, DINCH, DINP, DIDP, and DPHP). The distinction between the two groups was made according to the current legislation and the trends of plasticizers in use (REACH, 2018; Directive 76/769/EEC; EPA, 2016; Directive 2005/84/EC; EU, 2011; Bui et al., 2016; Brandon et al., 2015; Xie et al., 2015; Liang and Xu, 2014;). Manufacture or import volumes in the economic area of Europe are summarized in Table 1 for LPEs and APs. For that reason, we consider DINP and DIDP in the group of APs even if they are phthalates. We hypothesized that the type of the floor may affect the profile of indoor contamination, especially for the LPEs.

Thus, the main aims of the study were: (i) to investigate the concentration levels of the targeted compounds in indoor dust samples and if differences in the levels of plasticizers exist between countries (ii) to investigate the profile of contamination in various indoor environments and relationships to possible indoor sources, and (iii) to evaluate human exposure to the targeted plasticizers via dust ingestion and dermal absorption.

2. Materials and Methods

2.1 Chemicals and Reagents

Labeled dibenzyl phthalate (DBzP-d_4) was purchased from Accustandard (New Heaven, CT, USA and it was used as internal standard (IS) for the quantification of the targeted analytes. Chlorobiphenyl CB-207 was purchased from TCI Europe (Zwijndrecht, Belgium) and was used as recovery standard (RS). Standards of DMP, DEP, DNBP, DIBP, BBP, DEHP and DPP, DBA, ATEC, DBS, ATBC, DEHA, DPCP, BTHC, DEHT, THTM, TOTM, DINCH, DINP, DIDP, DPHP were purchased from Accustandard (New Heaven, CT, USA). Indoor dust standard reference material SRM 2585 was purchased from the US National Institute of Standards and Technology (NIST, Gaithersburg, MD, USA). Florisil® ENVI (500 mg, 3 mL) was purchased from Supelco

(Bellefonte, PA, USA). All solvents were chromatography grade. *n*-hexane (*n*-Hex) was purchased from Acros Organics (Belgium), ethyl acetate (EtAc), and iso-octane were purchased from Merck (Darmstadt, Germany).

2.2 Sample collection

Dust samples were collected from the interior of 33 homes from Belgium (*n=18, February 2017)*, Ireland *(n=6, January 2017)* and the Netherlands (*n=9, October 2017*); 16 offices divided between Sweden (*n=7, December 2016-February 2017)* and the Netherlands (*n=9, October 2017*); and 3 daycare centers from Sweden (*n=5 February-May 2016),* two of which were sampled in winter (February 2016) and in spring (May 2016), whereas the other one was sampled only during winter (Supplementary Material, Table S1). Dust sampling was conducted in most microenvironments using a vacuum cleaner equipped with a nylon sock (25 µm pore size) (Allied Filter Fabrics Pty Ltd, Australia). One m^2 of carpet was vacuumed for two minutes or four m^2 of bare floor for four minutes (Harrad et al., 2008). To avoid cross contamination, one nylon sock was used per sample and the vacuum cleaner tools were thoroughly rinsed with ethanol after each sampling. After collection, the socks were firmly folded, sealed in zip-lock plastic bags and carried to the lab. Dust samples were stored at -20°C pending analysis and then sieved (500µm) in room temperature and immediately extracted,

For daycare centers in Sweden, the sampling method was similar to that described by Sahlstrom et al (2012) and Thuresson et al (2012). The nozzle (polypropylene) and an inserted metal filter (acid-proof and stainless steel according to Swedish standard SS 2443, pore size 500 µm) were rinsed with ethanol (96%) after each sampling (no ultrasonic bath) and mounted on a vacuum cleaner. Cellulose filters in styrene-acrylonitrile holders were used and samples were collected above floor level, but not limited to the 1 m-level. Textile surfaces (like matrasses and sofas etc.) were not vacuumed. There was no time-keeping during sampling. Samples were stored in a freezer (-20° C) pending analysis.

2.3 Analytical method

Extraction and clean-up

For the targeted plasticizers, the analytical method used was based on existing protocols (Christia et al., 2018; Bergh et al., 2011) with slight modifications. LPEs and APs were simultaneously extracted from dust samples. Dust aliquots of 20 mg were weighed in pre-cleaned glass tubes (solvent washed and baked at 400°C) and spiked with 50 µL of IS D_4-DBzP (20 ng/µL). Samples were extracted using 2.5 mL of *n*-Hex/acetone (1:1, *v/v*) by a combination of vortexing and ultrasonication (2 x 1 min of vortex and 10 min of ultrasonication) repeated two times. After each extraction cycle, extracts were centrifuged at 3500 rpm for 3 minutes. Supernatants were then collected and transferred into clean glass tubes. The pooled

supernatants were evaporated to near dryness under a gentle nitrogen stream, redissolved in 1 mL of *n*-Hex and vortexed for 1 minute. Florisil® ENVI cartridges (500 mg, 3 mL) were prewashed with 6 mL of EtAc and 6 ml of *n*-Hex. The extracts were quantitatively transferred into the cartridges and fractionation was achieved by eluting the first fraction (F1) with 12 mL of *n*-Hex and the second fraction (F2) with 10 mL of EtAc. F2 was evaporated to near dryness under a gentle nitrogen stream, whereas F1 was discarded. After evaporation, F2 was dissolved in 50 µL of iso-octane and 50 µL of RS CB-207 (50 pg/µL) and transferred to an amber injection vial for further GC-EI-MS analysis.

2.4 Instrumental Analysis

Target compounds were analyzed using an Agilent GC coupled to an Agilent 5973 MS operated in electron ionization mode (EI). The GC system was equipped with a HT-8 column (25 m x 0.22 mm, 0.25 µm) electronic pressure control and a programmable-temperature vaporizer (PTV) inlet. The injection temperature was set at 90 °C, held for 0.04 min, ramped at 700 °C/min to 300 °C, held 25 min. Injection (1 µL) was performed under a pressure of 14.36 psi until 1.25 min and purge flow to split vent of 50 mL/min after 1.25 min, ramped at 15 °C/min to 200 °C, held for 3 min, ramped at 5 °C/min to 270 °C, ramped at 20 °C min^{-1} to 310 °C, held 12 min. Helium was used as a carrier gas with a flow rate of 1.0 mL/min until 28 min, then increased to 1.5 mL/min. The mass spectrometer was run in SIM mode with 2 characteristic ions acquired for each analyte and the IS (details are reported in Table S2 & Fig. S2).

2.5 Quality Assurance & Quality Control

The quality assurance and control were performed by analyzing SRM 2585 for the targeted compounds. This SRM is not certified for plasticizers but has been analyzed for them in other studies and results reported in the literature. The accuracy ranged between 58 and 89% compared to the mean values reported in literature (Larsson et al., 2017; Luongo & Ostman, 2016; Mercier et al. 2014, Bergh et al., 2012) (Table S3). Only DEHP values were slightly underestimated possibly due to its ubiquitous presence in the procedural blank that were subtracted from the real samples.

In total, ten procedural blanks (two per batch) were analyzed to check for laboratory contamination. The mean blank concentrations of target compounds were subtracted from the values found in the real samples and SRM. Limits of quantification (LOQs) were defined as follows; (i) for each compound that was present in the blank samples, a value equal to the mean blank concentration plus 3 times the standard deviation (SD) was used as the LOQ, (ii) for compounds that were not present in the blank samples, the LOQ was based on a signal/noise ratio of 10 (S/N=10). The LOQ range for the LPEs was 0.1 to 2 µg/g and for APs from 0.2 to 1 µg/g, except for DINCH, DINP and DIDP, where the LOQ was 5 µg/g. Concentration values below LOQs were treated as LOQ*f during statistical analyses, where f is the detection frequency of the compound above the LOQ in the samples. Mean recovery of the IS was 114 ± 16%.

2.6 Estimation of daily intake and potential health risk

The estimation of the daily intake was based on the approach previously reported by Christia et al. (2017) and modified according to the sampled indoor environments. The average daily dose (ADD) of plasticizers via dust ingestion and dermal absorption and the hazard quotient (HQ) were calculated using the following equations:

$$ADD_{ingestion} = C_{dust} \times IngR \times EF \times ED / (BW \times AT) \quad (1)$$

$$ADD_{dermal} = C_{dust} \times SA \times DA \times AF \times EF \times ED / (BW \times AT) \quad (2)$$

$$HQ = ADD_{ingestion} / RfD \quad (3)$$

where C_{dust} is the concentration of the compound detected in dust (μg/g); IngR is the daily ingestion rate of dust (30 mg/day for adults and 60 mg/day for toddlers (USEPA 2014; Wang et al., 2013; Kang et al., 2011; Kurt-Karakus et al., 2012); EF is the exposure frequency over one year (347 days; 95th percentile of 365 days; USEPA, 1989); ED is the life-time exposure duration (30 years for adults; 2 years for toddlers; USEPA, 2001); BW is the body weight (70 kg for adults, 16 kg for toddlers; USEPA, 1989); AT is the averaging time; SA is the exposed body surface area (assumed 4,615 cm^2 for adults and 2,564 cm^2 for toddlers, (Abdallah et al., 2015); DA is the dust adhered to skin (0.01mg/cm^2 for adults and 0.04 mg cm^2 for toddlers, (Abdallah et al., 2015); AF is the fraction of contaminant adsorbed by skin (0.00048 for DMP, 0.01025 for DEP, 0.0006 for DIBP, 0.00078 for DNBP, 0.00035 for BBzP, 0.000053 for DEHP, 0.000031 for DINCH and DINP, 0.000039 for DIDP and DPHP; Giovanoulis et al., 2018) and RfD is the reference dose for each compound (EPA, 1993). The RfD parameter is an estimate of daily exposure of the human population, including sensitive sub-population groups (i.e. toddlers) that is likely to be without an appreciable risk of deleterious non-cancer effects during a lifetime (EPA, 1993). RfDs were calculated and/or derived from the literature for each compound (Table S4). For those compounds for which both the calculated and the literature RfD value were available, the lower value was selected in equation (3) to represent the stricter limit. The calculated RfDs were derived from the following equation from the USEPA (2002):

$$RfD = NOAEL / UF \quad (4)$$

where NOAEL is the no-observed-adverse-effect level from chronic exposure (mg/kg/day) and UF is an uncertainty factor (1000).

2.7 Statistical analysis

Descriptive statistics were computed using IBM SPSS version 24.0.0.0. T-test values higher than 0.5 (t>0.5) were accepted as statistically significant. Correlation coefficients were calculated using the concentrations of individual and total plasticizers relative to characteristics of the indoor environment (e.g. building

construction year, type of furnishing, number of electronics, etc.). P-values lower than 0.05 ($p<0.05$) were considered statistically significant. Pearson correlation factors were calculated to evaluate a possible relationship between the detected concentrations of individual and total plasticizers and the type of floor (PVC and non-PVC).

3. Results and discussion

3.1 Targeted plasticizers in indoor dust

For the analyzed floor dust samples, 17 of 21 targeted plasticizers were found in concentrations above the LOQs (Tables S6-S11). DMP, ATEC, DPP and THTM were not detected in most of the samples (Table 2). Floor dust from Swedish offices and above-floor dust from the daycare centers sampled during the winter had the highest concentrations of $\Sigma_{plasticizers}$ of all microenvironments, with mean concentrations of 1,800 and 1,200 µg/g, respectively. For the two Swedish daycare centers that were sampled in both seasons, lower concentrations were found in the spring samples which could be due to the open windows as a better ventilation.. However, the sample size is too small to draw firm conclusions. The lowest mean concentration of total plasticizers was found in Belgian homes, 335 µg/g. The mean percent contribution of each compound group to the total plasticizer concentration was about 60% for the APs and about 40% for the LPEs for all indoor environments (Fig. S3).

The mean contamination pattern for each type of microenvironment and country was derived from the calculated percentage contribution for each individual plasticizer to the total concentration. The domestic environments of Belgium (BE), Ireland (IRE) and the Netherlands (NL) showed a similar pattern (Fig S4), with DEHP, DEHT, and DINP being the predominant plasticizers in home dust. The intracountry variability of the environments was not significant except for the Swedish offices where 3 out of 7 offices showed higher concentrations. In home environments, the main contributor for PLEs was DEHP (27% BE, 25% IRE and 36% NL) and for APs, the main contributor was DEHT (18% BE, 28% IRE and 21% NL). The office environments showed more diverse patterns compared to homes. DEHP (37%) and DIDP (36%) contributed most in Swedish (SE) office floor dusts, together reaching almost 75% of the total measured plasticizers. On the other hand, DEHP (38%) was the predominant LPE in NL offices, followed by DIDP (13%), DINP (14%) and DEHT (11%). The contribution of plasticizers to Swedish (SE) preschool above-floor dust was similar between the two sampling seasons, with DEHP (20% in winter, 17% in spring), DINP (33% in winter, 40% in spring) and DIDP (14% in winter, 22% in spring) being the major measured compounds. Higher concentrations of individual compounds were found in samples collected during the winter sampling.

Compared to previously published studies for plasticizers in dust from different indoor environments, the BE (335 µg/g) and NL homes (410 µg/g) in this study were the least contaminated domestic environments. Floor dust from IRE homes (540 µg/g) showed similar total plasticizers levels as from homes in the USA (490 µg/g & 464 µg/g) (Subedi et al., 2017; Bi et al., 2015), but lower levels than from above-floor surface dust in SE homes (1,200 µg/g) (Bergh et al., 2011). The concentration levels in the present

home samples from the three countries (BE, IRE and NL) were also lower than those found in floor dust collected in Norwegian homes (Giovanoulis et al., 2018).

Concentration levels of plasticizers detected in the SE offices (1,800 μg/g) from the current study were the same (1,800 μg/g) as those reported by Bergh et al. (2011) for above-floor surface dust from ten Swedish offices collected in 2006. However, US office dust had lower total concentrations (2,471 μg/g). The SE daycare center dusts from the current study had lower levels (1,200 (winter) and 670 μg/g (summer)) compared to US daycare centers (2,153 μg/g) (Subedi et al., 2017) and above-floor surface dust from SE daycare centers collected in 2006 (2,300 μg/g) (Bergh et al., 2011). The total concentration levels were determined by the number of the target analytes per study but for all studies higher concentrations of all targeted compounds were found in floor dust from non-domestic environments than in homes.

3.2 Source profiling

Correlations between the concentrations of the total plasticizers, dominant compounds and indoor characteristics like number of furniture and electronics, building age or year of renovation were not statistically significant ($p>0.05$) (Table S5). However, as collection of floor dust was the sampling method used for most samples, we hypothesized that the type of sampled surface could be a significant parameter affecting the contamination profile of the dust.

Fig. 1 shows a comparison of the plasticizer concentrations in floor dust collected from microenvironments with or without PVC floors. Higher total plasticizer concentrations were found in floor dust collected from PVC floors ($p<0.01$). The link of floor type exists between high concentrations of the analytes and the non-domestic environments. PVC floors are mostly placed in public buildings with large floor area and high daily use, where durability and less need for maintenance are essential (PVC; Plasticizers, Information Center, 2015). Since offices were equipped with this type of floor, it could be a main source contributing to the high levels of plasticizers. On the other hand, domestic environments are usually equipped with wooden (parquet) floors, carpets, or ceramic tiles, which are more expensive materials and of higher quality. Nevertheless, floor dust is a complicated matrix, since it can be easily affected by the material of the floor, cleaning products and walking/cleaning frequency.

Non-PVC floors (e.g., parquet, tiles, laminate, or carpets) were present in 39 out of the 52 environments and PVC floors were in 13 interiors. Offices environments were equipped with PVC or linoleum floors whereas homes were equipped mainly with parquet and tiles floor. The concentration of total plasticizers may be relevant with the floor type. As individual compounds, DINP, DIDP, DNBP, DPHP and DEHA showed rather high relevance with the floor type as well. Excluding DEHA, which is an AP, the other compounds were previously used in PVC flooring, but have been phased out by the European industry (ECHA, 2012). However, it is under dispute whether there is complete compliance with the relevant legislation by producers and importers (ECHA, 2012; Sackmann et al., 2018). Additionally, the presence of these plasticizers in dust could be due to the presence of old PVC flooring, recycled materials in PVC flooring

and/or their use in other indoor equipment that could act as potential sources of contamination (ECHA, 2012). Additionally, a co-occurrence of DEHP, DINP, DIDP, DPHP, DNBP, DIBP, BBzP could suggest that these compounds may derive from the same source, likely the floor. LPEs (DEHP, DNBP, DIBP, and BBzP) were internally positively correlated and also positively correlated with several APs (DINP, DIDP, and DEHA) which suggests similar sources ($p<0.05$).

DEHP was the major compound found in floor dust and its predominant presence in several indoor environments, such as homes, offices, daycare centers and salons, has already been confirmed by numerous studies in the literature (Wang et al., 2017, Subedi et al., 2017, Larsson et al., 2017, Fromme et al., 2016; 2013; 2004, Bi et al., 2015, Luongo & Ostman 2016, Gaspar et al., 2014, Gevao et al., 2013, Kubwabo et al., 2013, Guo & Kannan 2011, Nagorka et al., 2011, Bergh et al., 2011, Langer et al., 2010, Abb et al., 2009, Kolarik et al., 2008, Bornehag et al., 2005, Morgan et al., 2004). Being a general-purpose plasticizer, it is possibly released from multiple indoor sources (e.g. building construction materials, PVC flooring, furniture, toys, shoe soles, leather products, food packaging and/or storage products, electronic equipment, etc.) (ECHA, 2018). DEHP, DINP and DIDP found highly relevant with the presence of PVC flooring in indoor environments. DINP was present in all dust samples but the highest contribution was found in SE offices (36%) and SE daycare centers (33% in winter, 40% in spring), which were environments that all had PVC flooring. DINP is also incorporated in wires of electronics (American Chemistry Council, 2012), but no correlation with electronic equipment counts was found. For DIDP, the highest contribution to total plasticizer concentrations was estimated for NL offices (13%) and SE daycare centers (14% in winter and 22% in spring), most probably due to its presence in the floor type ($p<0.01$). DIDP is also a major plasticizer for cable and wire applications in Europe (ECPI, 2011). It is characterized by lower vapor pressure and higher permanency than DINP (BASF, 2011). However, its higher cost leads to optional application in products where advance technical characteristics are needed (ECHA, 2012).

3.3 Human exposure via dust ingestion and dermal absorption

To assess the potential health risk due to plasticizer intake, a non-carcinogenic risk assessment was conducted using the calculated HQs (eq. 3) (Table S6). The available RfDs for plasticizers are provided in Table S4. Figures 2, 3, and 4 represent the $ADD_{ingestion}$ values, whereas ADD_{dermal} values were included in Tables S13- S15. Adults in BE homes were found to be the least exposed population of the study for all the plasticizers, whereas adults in SE offices were the most exposed especially to DEHP and DINP. Toddlers were more exposed to indoor contamination in SE daycare centers and in NL homes, possibly related to the floor type. The mean $ADD_{ingestion}$ values for adults were 0.02 μg/kg/day (BE homes), 7 μg/kg/day (IRE homes), 9 μg/kg/day (NL homes), 23 μg/kg/day (SE offices), 7 μg/kg/day (NL offices) and 13 μg/kg/day (SE daycare centers). The same values for toddlers were 1 μg/kg/day (BE homes), 4 μg/kg/day (IRE homes), 7 μg/kg/day (NL homes) and 8 μg/kg/day (SE daycare centers). The ADD_{dermal} values for all the environments were several orders of magnitude lower than the ones of dust ingestion for adults and toddlers.

Major contributors to human exposure were DEHP, DINP and DIDP for LPEs and DEHT for APs. None of the HQs calculated in Table S12 was higher than 1, indicating an unlikely health risk *via* dust ingestion. Nevertheless, it is crucial to take into consideration that dust ingestion represents only one human exposure pathway to plasticizers and it is not estimated based on the actual time fraction per day that a person spends in each microenvironment since that kind of information was not available. Additionally, this study did not provide sufficient data to estimate human exposure to plasticizers through food intake and air inhalation (Giovanoulis et al., 2018; Miao et al., 2017; Schecter et al. 2013; Fierens et al., 2012). Especially for DEHP, food intake has been estimated as the most important exposure pathway (Giovanoulis et al., 2018; Sioen et al., 2012; Wittassek et al., 2011; Wormuth et al., 2006). The indicative values of average daily dietary intake (EDI) for LPEs are lower than those reported for dust ingestion (e.g. 0.092- 1.60 μg/kg bw/day and 0.041-3.55 μg/kg bw/day) but the intake frequency is higher for food consumption than dust ingestion (Giovanoulis et al., 2018; Guo et al., 2012; Sioen et al., 2012).

4. Conclusions

LPEs and APs were detected in floor dust from all studied indoor environments. Higher concentrations in dust were found for APs than LPEs, possibly indicating the gradual replacement of the LPEs with a new generation of replacement plasticizers. The major LPE found was DEHP, whereas for APs, the predominant compounds were DIDP, DINP and DEHT. A difference was found with higher levels of plasticizers in dust collected from PVC floors, indicating that the flooring material could be a main source of contamination for the collected dust. As PVC flooring may be used in offices, daycare centers and public places to a larger extent, these microenvironments may be more contaminated than homes. No correlation was observed between the concentrations of the analytes and other characteristics of the indoor environments (e.g. furniture, electronic equipment, year of building construction, year of renovation etc.). Human exposure to plasticizers from dust ingestion was found to be higher in non-domestic microenvironments due to their higher concentrations of plasticizers. Based on intake from dust ingestion and dermal absorption, the hazard quotients for each compound were much lower than 1, indicating negligible health risk from this exposure pathway.

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Table 1. European production/ import volumes for the target plasticizers.

Compound	Manufacture/Import volume (tone/year)	References
LPEs		
DINP	100,000-1,000,000	*ECHA, 2017*
DIDP	100,000-1,000,000	*Danish EPA, 2013*
DEHP	10,000-100,000	*ECHA, 2018*
DMP	1,000-10,000	*ECHA, 2017*
DNBP	1,000-10,000	*ECHA, 2017*
DIBP	1,000-10,000	*ECHA, 2017*
BBzP	1-10	*ECHA, 2017*
APs		
DEHT	100,000-1,000,000	*ECHA, 2017*
DPHP	100,000-1,000,000	*ECHA, 2017*
DINCH	~200,000	*Bui et al., 2016*
ATBC	10,000-100,000	*ECHA, 2017*
TOTM	10,000-100,000	*ECHA, 2017*
DEHA	10,000-100,000	*ECHA, 2017*
DBA	100-1,000	*ECHA, 2017*
DBS	100-1,000	*ECHA, 2017*
ATEC	10-100	*ECHA, 2017*

Table 2. Descriptive statistics for each indoor environment (μg/g).

	BE homes (n=18)					***IRE homes (n=6)***				
	Mean	*Median*	*SD*	*Min*	*Max*	*Mean*	*Median*	*SD*	*Min*	*Max*
LPEs										
DEP	0.42	1.4	0.38	0.16	1.5	1.9	0.64	2.4	0.39	6.6
DIBP	7.6	9.4	12	1.2	51	32	7.0	58	4.6	150
DNBP	14	22	26	0.67	109	37	7.1	74	5.7	187
BBP	4.3	8.5	5	0.20	16	3.9	3.9	1.7	2.0	6.4
DEHP	88	62	115	9.0	497	127	114	80	24	254
APs										
DBA	0.33	<LOQ	0.42	<LOQ	1.5	0.23	<LOQ	0.22	<LOQ	0.60
DBS	<LOQ	<LOQ	<LOQ	<LOQ	0.61	<LOQ	<LOQ	0.34	<LOQ	0.84
ATBC	12	1.9	27	0.22	101	8.9	7.5	4.6	4.8	17
DEHA	19	5.4	65	0.27	272	3.0	2.9	1.2	1.6	4.7
DPCP	0.19	<LOQ	0.61	<LOQ	2.5	0.78	0.40	1.2	<LOQ	3.1
BTHC	0.76	0.31	2.6	<LOQ	11	0.25	0.23	<LOQ	<LOQ	0.46

DEHT	36	23	27	5.1	101	151	164	104	32	247
TOTM	10	5.2	30	0.46	130	2.3	2.6	0.88	1.2	3.2
DINCH	73	74	245	1.2	1051	38	33	39	1.7	111
DINP	52	26	67	5.2	296	84	72	27	62	121
DIDP	13	10	16	4.4	59	30	26	19	16	67
DPHP	3.4	3.9	3.8	0.90	16.1	14	7.7	18	2.5	49
ΣPlasticizers	***335***					***540***				

	NL homes (n=9)					***NL offices (n=9)***				
	Mean	Median	SD	Min	Max	Mean	Median	SD	Min	Max
LPEs										
DMP	<LOQ	<LOQ	<LOQ	<LOQ	0.31	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
DEP	0.71	0.23	1.2	0.19	3.82	0.35	0.34	<LOQ	0.20	0.53
DIBP	7.0	4.7	7.9	<LOQ	26	5.6	3.2	6.5	1.3	22
DNBP	20	3.2	47	1.2	146	45	6.0	83	2.3	230
BBP	5.7	2.7	6.8	0.70	18	11	5.3	14	0.70	36
APs										
DEHP	123	111	87	32	307	240	150	201	31	651
DBA	<LOQ	<LOQ	<LOQ	<LOQ	0.20	<LOQ	<LOQ	<LOQ	<LOQ	0.50
DBS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.23	<LOQ	0.31	<LOQ	0.79
ATBC	6.0	2.3	7.6	0.20	21	6.1	4.7	5.7	0.40	17
DEHA	4.7	2.4	6.0	0.20	17	14	6.4	16	1.1	46
DPCP	0.10	<LOQ	0.26	<LOQ	0.80	0.59	0.30	0.56	<LOQ	1.5
BTHC	<LOQ	<LOQ	<LOQ	<LOQ	0.64	0.49	0.47	0.33	<LOQ	1.0
DEHT	132	37.1	241	6.9	764	57	21	75	8.5	229
TOTM	9.8	1.9	16	<LOQ	46	11	5.4	15	0.6	46
DINCH	10	9.5	6.1	<LOQ	19	28	12	44	7.0	144
DINP	59	34	49	<LOQ	152	81	56	82	38.8	297
DIDP	18	13	18	<LOQ	62	60	65	43	11	147
DPHP	16	4.3	29	<LOQ	90	9.0	5.1	9.0	1.2	24
ΣPlasticizers	***410***					***570***				

	SE offices (n=7)				
	Mean	Median	SD	Min	Max
LPEs					
DMP	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
DEP	0.61	0.59	0.28	0.28	0.98
DIBP	6.3	3.5	6.8	0.20	21
DNBP	57	25	60	6.8	176
BBP	11	8.3	7.9	4.0	25
DEHP	751	200	786	40	1957
DPP	2.5	<LOQ	6.6	<LOQ	17
APs					
DBA	<LOQ	<LOQ	<LOQ	<LOQ	0.20
ATEC	0.59	<LOQ	1.6	<LOQ	4.2
DBS	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
ATBC	35	2.3	56.	0.54	119
DEHA	46	4.8	102	2.8	276
DPCP	0.29	0.10	0.24	0.10	0.60

BTHC	0.35	0.30	0.31	<LOQ	0.92
DEHT	48	19	47	13	136
TOTM	9.1	1.6	14	0.72	39
DINCH	25	21	20	7.5	66
DINP	648	287	656	157	1872
DIDP	134	65	164	22	485
DPHP	15	10.3	14	3.4	46
ΣPlasticizers	***1800***				

	SE daycare centers-Winter sampling (n=3)					***SE daycare centers-Spring sampling (n=2)***				
	Mean	Median	SD	Min	Max	Mean	Median	SD	Min	Max
LPEs										
DMP	2.0	0.50	3.1	<LOQ	5.6	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
DEP	2.0	1.1	2.1	0.33	4.4	0.28	0.28	0.18	0.15	0.40
DIBP	20	19	15	5.6	35	6.8	6.8	0.32	654	7.0
DNBP	86	62	64	37	158	21	21	7.8	15	26
BBP	104	78	88	32	201	29	29	25	11	46
DEHP	252	200	152	133	423	109	109	37	83	140
APs										
DBA	0.77	0.60	0.29	0.60	1.1	0.25	0.25	0.07	0.20	0.30
DBS	0.09	<LOQ	0.16	<LOQ	0.28	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
ATBC	43	32	42	8	89	6.7	6.7	3.7	4.2	9.3
DEHA	26	4.8	38	3.3	70	2.2	2.2	0.33	1.9	2.4
DPCP	0.77	0.10	1.2	0.10	2.1	0.10	0.10	<LOQ	0.10	0.10
BTHC	0.68	0.59	0.20	0.54	0.91	0.23	0.23	0.19	0.10	0.37
DEHT	121	83	102	43	237	57	57	21.4	42	72
TOTM	1.8	1.48	1.2	0.82	3.1	0.96	0.96	0.08	0.91	1.0
DINCH	31	22	18	18	52	16	16	9.6	9.3	23
DINP	372	239	280	182	694	289	289	313	68	511
DIDP	142	151	82	55	219	131	131	124	43	219
DPHP	3.5	3.9	1.4	1.97	4.7	3.7	3.7	1.6	2.6	4.8
ΣPlasticizers	***1200***					***670***				

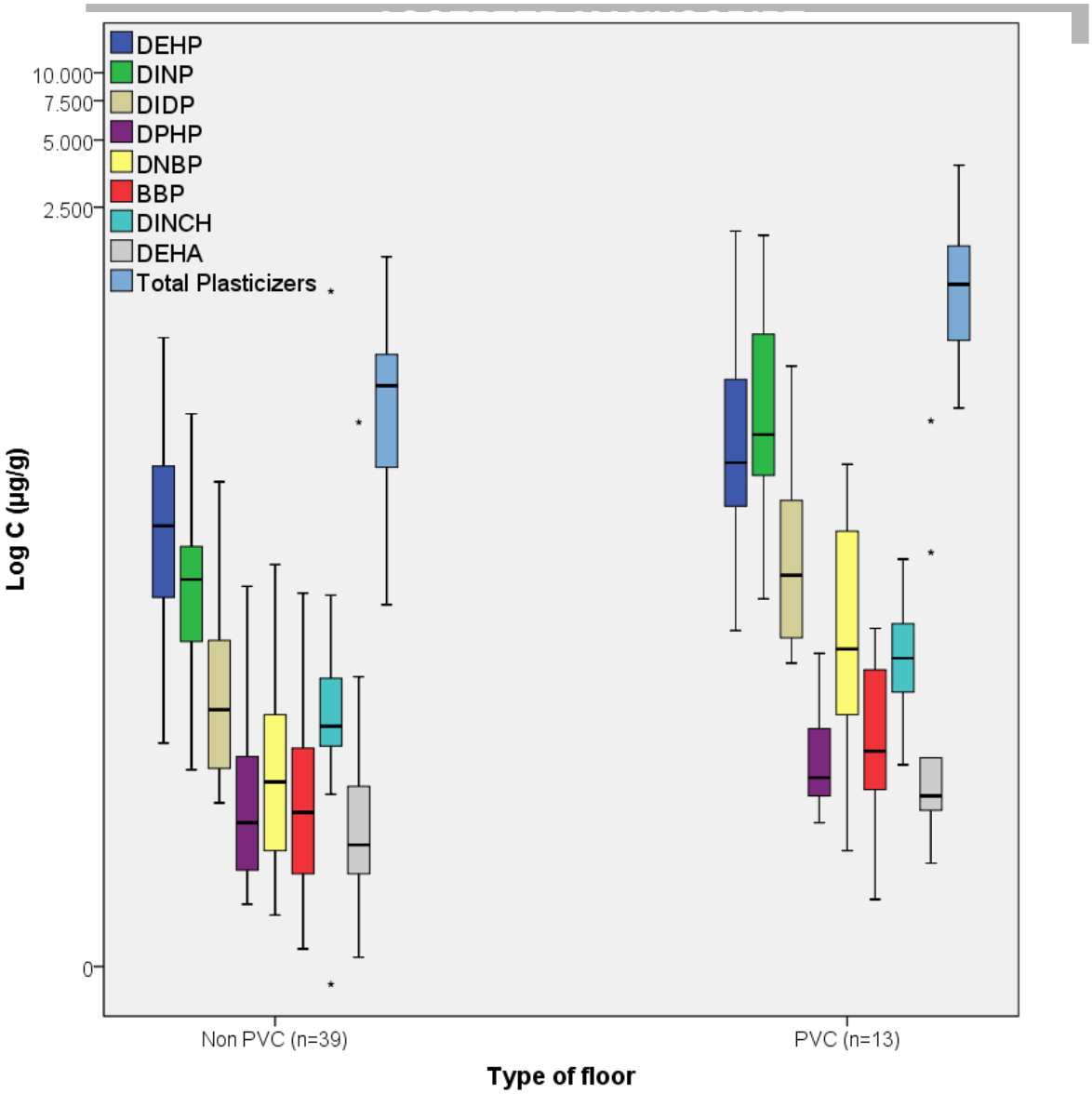

Figure 1. Box-whisker comparison of concentrations of individual & total plasticizers (µg/g) in sampled dust from PVC and non-PVC floors. The horizontal line is the (median), the box represents the total concentration of plasticizers and the whiskers represent the lowest and the highest value of total plasticizers. The * indicates the extreme values.

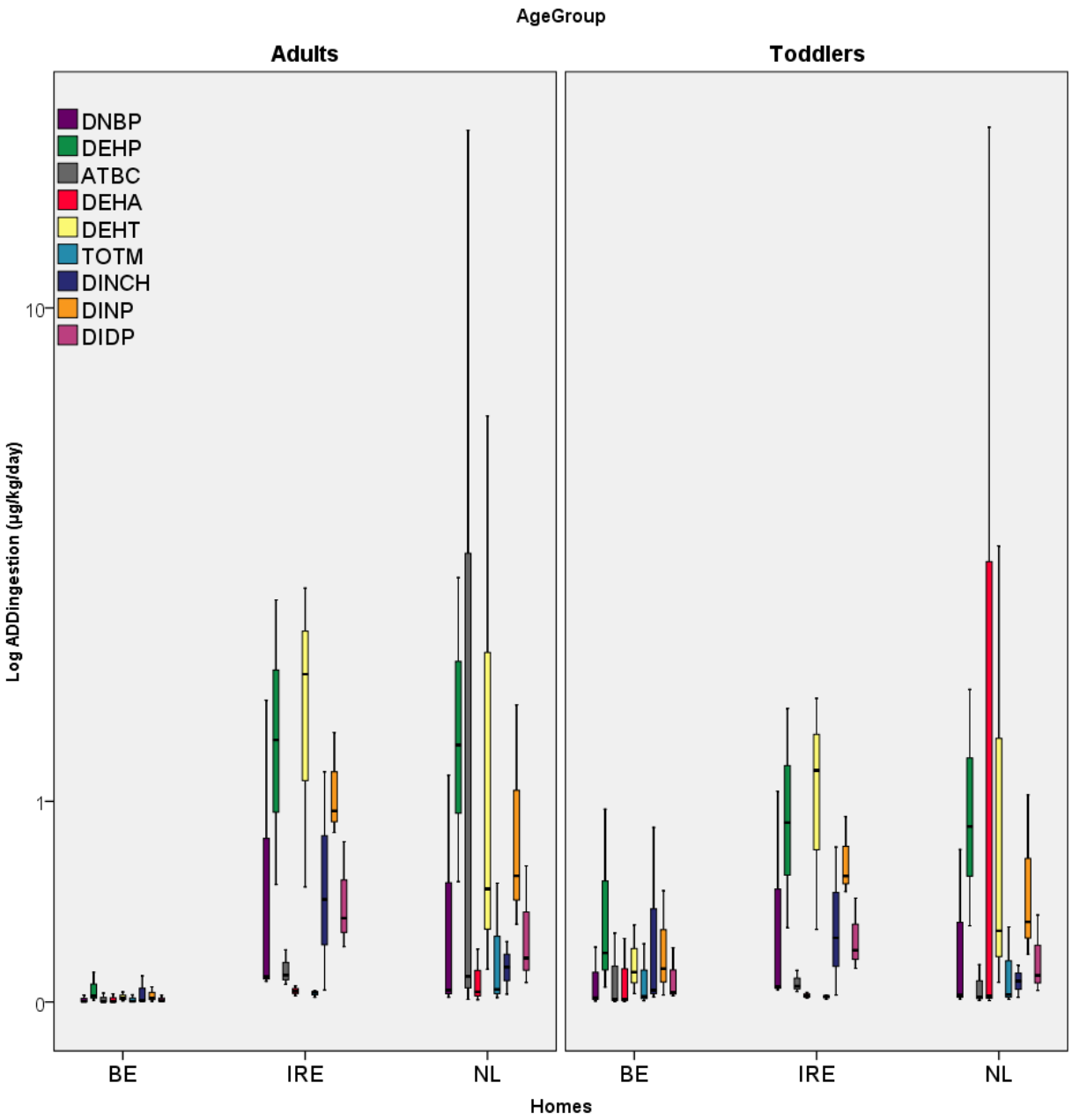

Figure 2. $ADD_{ingestion}$ values for plasticizers (µg/kg/day) in homes.

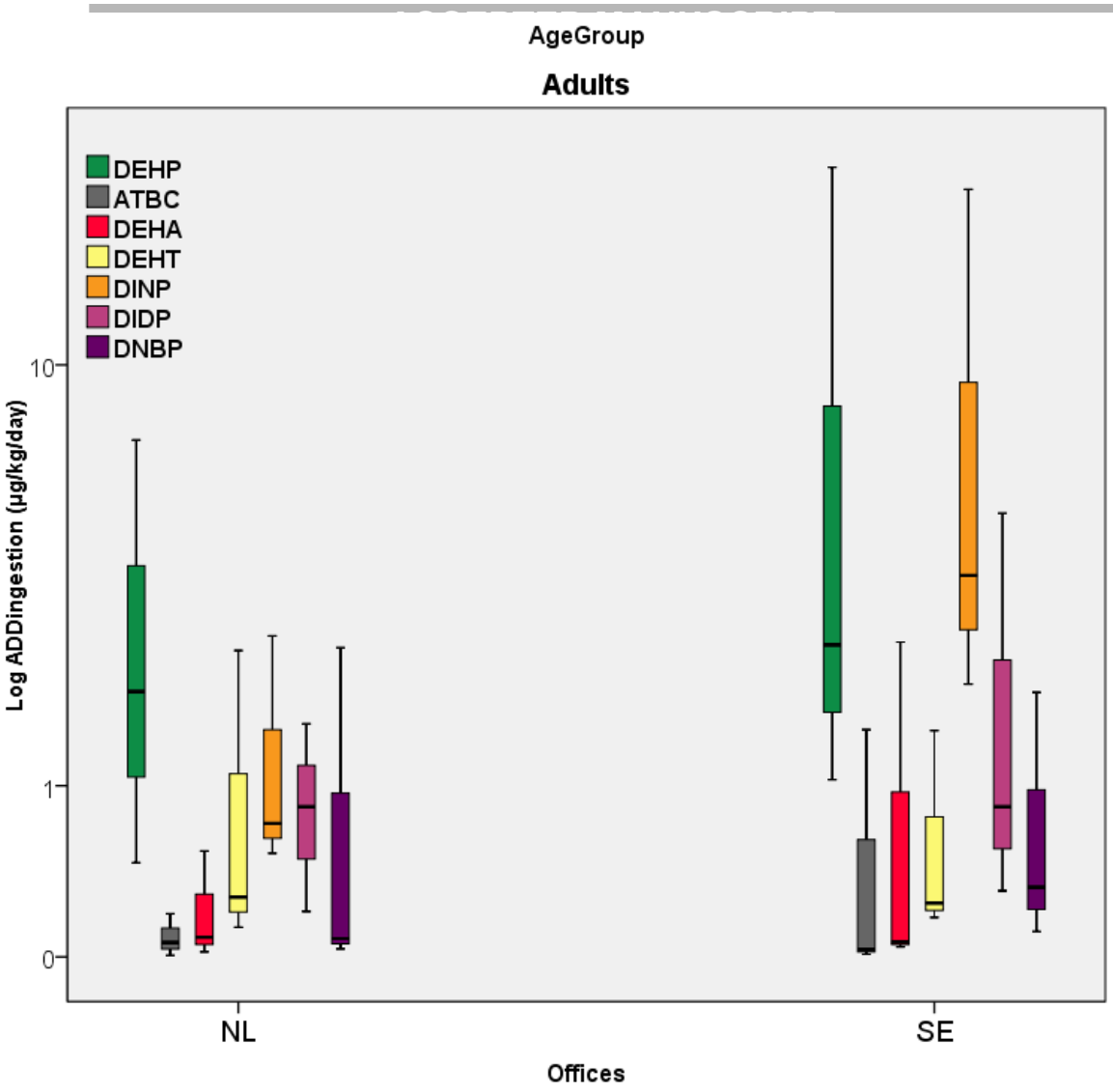

Figure 3. $ADD_{ingestion}$ values for plasticizers (µg/kg/day) in offices.

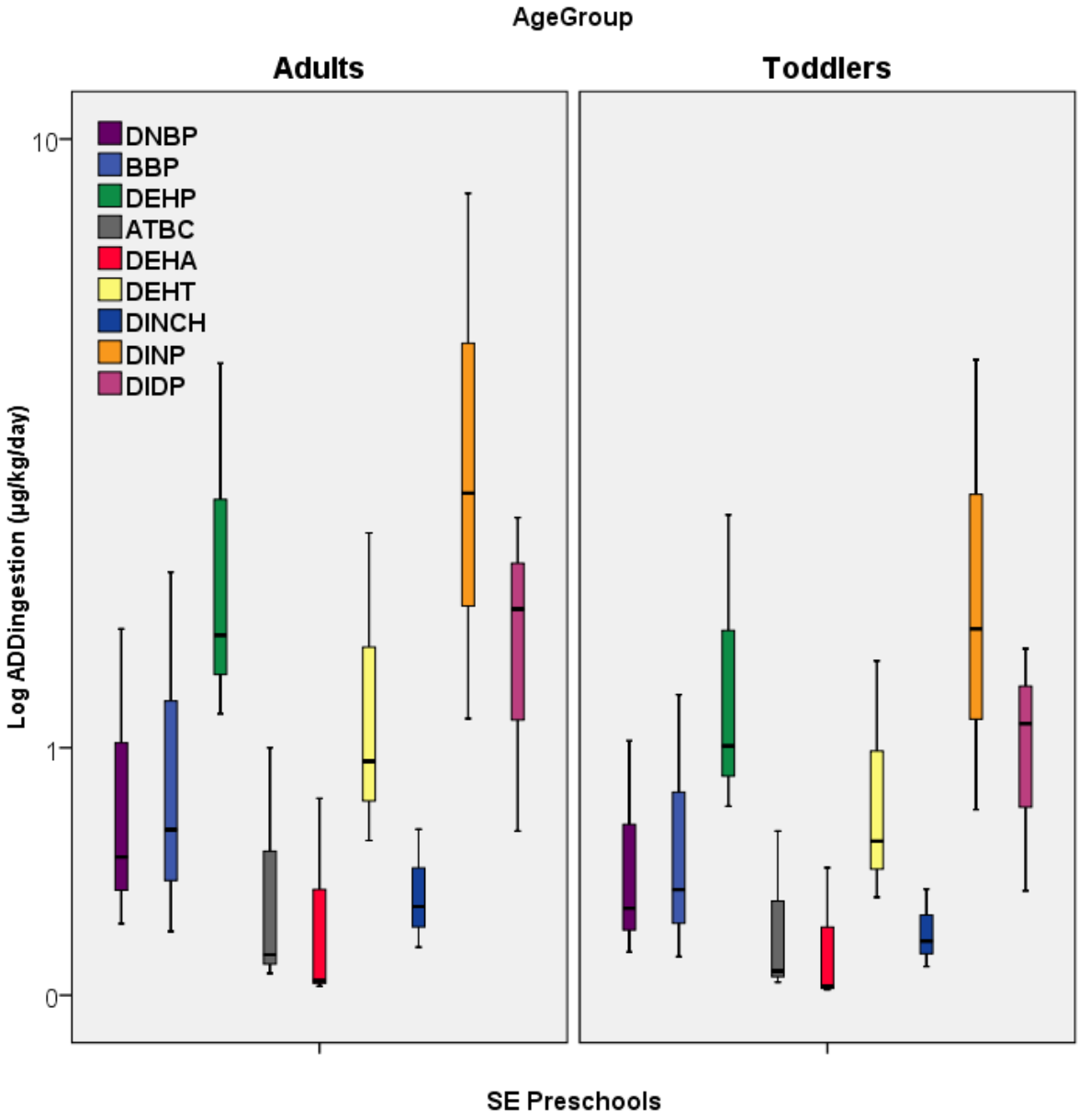

Figure 4. $ADD_{ingestion}$ values for plasticizers (µg/kg/day) in daycare centers.

Highlights

- Higher plasticizer levels were found in dust from ''non-domestic'' environments.
- DEHP was the dominant plasticizer even though its use has been restricted.
- DINP and DEHT were the dominant alternative plasticizers.
- The concentration of plasticizers can be related to the type of floor.
- Higher levels of APs in dust are an indication of the gradual substitution of LPEs.