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Phthalate and non-phthalate plasticizers in indoor dust from childcare facilities, salons, and homes across the USA☆

Bikram Subedi [a,*], Kenneth D. Sullivan [a], Birendra Dhungana [b]

[a] Department of Chemistry, Murray State University, Murray, KY, 42071, United States
[b] Department of Environmental Science, Baylor University, Waco, TX, 76798, United States

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ABSTRACT

The quality of indoor environment has received considerable attention owing to the declining outdoor human activities and the associated public health issues. The prolonged exposure of children in childcare facilities or the occupational exposure of adults to indoor environmental triggers can be a culprit of the pathophysiology of several commonly observed idiopathic syndromes. In this study, concentrations of potentially toxic plasticizers (phthalates as well as non-phthalates) were investigated in 28 dust samples collected from three different indoor environments across the USA. The mean concentrations of non-phthalate plasticizers [acetyl tri-*n*-butyl citrate (ATBC), di-(2-ethylhexyl) adipate (DEHA), and di-isobutyl adipate (DIBA)] were found at 0.51–880 μg/g for the first time in indoor dust samples from childcare facilities, homes, and salons across the USA. The observed concentrations of these replacement non-phthalate plasticizer were as high as di-(2-ethylhexyl) phthalate, the most frequently detected phthalate plasticizer at highest concentration worldwide, in most of indoor dust samples. The estimated daily intakes of total phthalates (n = 7) by children and toddlers through indoor dust in childcare facilities were 1.6 times higher than the non-phthalate plasticizers (n = 3), whereas estimated daily intake of total non-phthalates for all age groups at homes were 1.9 times higher than the phthalate plasticizers. This study reveals, for the first time, a more elevated (~3 folds) occupational intake of phthalate and non-phthalate plasticizers through the indoor dust at salons (214 and 285 ng/kg-bw/day, respectively) than at homes in the USA.

1. Introduction

Phthalates, esters of phthalic acids, have been used as plasticizers in diverse applications including children's toys, food containers, personal care products, medical devices, electronics, PVC floorings, and building materials at a single percent to tens of percent levels (Ma et al., 2014). Phthalates are semi-volatile organic compounds and do not chemically bound to the host polymer, therefore, are prone to leach from the plastic materials. The leached phthalates partition to the indoor air (log *K*oa = 6.70–12.56) and indoor materials (log *K*oc = 1.68–5.27) including indoor dust (Table 1). A thin layer of dust on the impervious surface in indoor environment was found to accumulate several organic contaminants (Bi et al., 2015; Liu et al., 2003). Therefore, indoor dust can be a sink and a repository for many indoor environmental contaminants including phthalates (Butte and Heinzow, 2002).

The occurrence and fate of phthalates in the indoor environment depends on several factors including usage, leachability, the volume of indoor air, the exchange rate of air (Fromme et al., 2004), moisture content (Hsu et al., 2017), interior surface/material composition (Jeon et al., 2016), and indoor temperature (Bi et al., 2015; Jeon et al., 2016). For example, the absorbed phthalate concentration in cotton and polyester clothes was found ~3 fold higher at 30 °C than at 21 °C in a test house in Austin, TX (Bi et al., 2015). However, overall contamination profile of commonly observed phthalates in the environment varies across the world (Guo and Kannan, 2011; Kang et al., 2012). DEHP is the most frequently detected (~100%) phthalate in indoor dust, and is reportedly the most dominant phthalate contaminant in indoor dust in USA,

Abbreviations: ATBC, acetyl tri-*n*-butyl citrate; BBP, butyl benzyl phthalate; DBP, di-*n*-butyl phthalate; DEHA, di-(2-ethylhexyl) adipate; DEHP, di-(2-ethylhexyl) phthalate; DEP, diethyl phthalate; DIBA, di-isobutyl adipate; DIBP, di-isobutyl phthalate; DMP, dimethyl phthalate; DOP, di-*n*-octyl phthalate.

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* Corresponding author. Department of Chemistry, Murray State University, 1201 Jesse D. Jones Hall, Murray, KY, 42071, United States.
E-mail address: bsubedi@murraystate.edu (B. Subedi).

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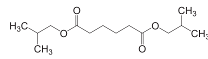
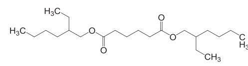
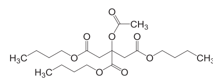
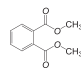
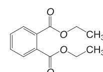
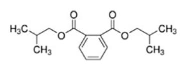
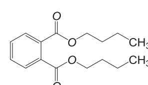
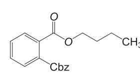
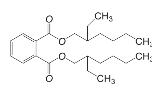
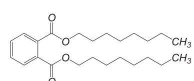
Table 1
Structures and physicochemical properties of target phthalates and non-phthalates plasticizers.

Analytes	Molecular structure	Molecular formula	Mol. Wt.	CAS	Water solubility[a]	Log K_{oa}[b]	Log K_{oc}[c]	Log K_{ow}[d]	VP (Pascal)[e]
Non-phthalate plasticizers									
di-iso-butyl adipate (DIBA) or hexanedioic acid, bis(2-methylpropyl) ester		$C_{14}H_{26}O_4$	258.4	141-04-8	5.649	7.850	3.1115	4.19	0.751
di-(2-ethylhexyl) adipate (DEHA) or di-octyl adipate		$C_{22}H_{42}O_4$	370.6	103-23-1	5.452E-04	12.871	5.2853	8.12	4.27×10^{-4}
acetyl tri-*n*-butyl citrate (ATBC) or (Citroflex A-4®)		$C_{20}H_{34}O_8$	402.5	77-90-7	0.6464	12.101	4.9428	4.29	6.07×10^{-4}
Phthalate plasticizers									
dimethyl phthalate (DMP)		$C_{10}H_{10}O_4$	194.2	131-11-3	2014	6.694	1.6789	1.66	0.263
diethyl phthalate (DEP)		$C_{12}H_{14}O_4$	222.2	84-66-2	287.2	7.023	2.1325	2.65	6.48×10^{-2}
di-iso-butyl phthalate (DIBP)		$C_{16}H_{22}O_4$	278.3	84-69-5	5.061	8.412	3.0673	4.46	4.73×10^{-3}
di-*n*-butyl phthalate (DBP)		$C_{16}H_{22}O_4$	278.3	84-74-2	2.351	8.631	3.2830	4.61	4.73×10^{-3}
benzyl-butyl phthalate (BBP)		$C_{19}H_{20}O_4$	312.6	85-68-7	0.9489	9.018	3.4102	4.84	2.49×10^{-3}
bis-(2-ethylhexyl) phthalate (DEHP)		$C_{24}H_{38}O_4$	390.6	117-81-7	1.132E-03	12.557	4.9977	8.39	2.52×10^{-5}
di-*n*-octyl phthalate (DOP)		$C_{24}H_{38}O_4$	390.6	117-84-0	4.236E-04	12.079	5.2743	8.54	2.52×10^{-5}

[a] Water solubility (mg/L, 255 °C) estimated from Log *K*ow using the US Environmental Protection Agency's EPISuite™, [WSKOWWIN v1.41].
[b] Log octanol-air partition coefficient (25 °C) estimated using the US Environmental Protection Agency's EPISuite™, [KOAWIN v1.10].
[c] Corrected Log soil adsorption coefficient estimated from Log K_{ow} values using the US Environmental Protection Agency's EPISuite™, [KOCWIN v2.00].
[d] Log octanol-water partition coefficient (25 °C) estimated using the US Environmental Protection Agency's EPISuite™, [KOWWIN v1.67].
[e] Vapor pressure for phthalate and non-phthalate plasticizers (Cousins and Mackay, 2000) and the US Environmental Protection Agency's EPISuite™, respectively.

Germany, and China (Fromme et al., 2004; Guo and Kannan, 2011; Kanazawa et al., 2010; Kang et al., 2012; Ma et al., 2014; Rudel et al., 2003). However, DBP (geometric mean concentration = 7860 μg/g) was the major phthalate contaminant in indoor dust in Bulgaria (Kolarik et al., 2008).

The contaminant in indoor dust including plasticizers can be an important marker of indoor exposure and public health (Ma et al., 2014; Mendell and Heath, 2005; Whitehead et al., 2011). The acute or chronic exposure of dialkyl *ortho*-phthalates can cause several adverse effects on human health including irreversible changes in the reproductive system (Kay et al., 2013, 2014) and cognitive performance in school children (Hutter et al., 2013). Exposure of plasticizers can be particularly critical for children due to the developmental juncture and a higher daily intake per unit body mass than adults due to children's higher hand-mouth activities. In addition, an elevated level (2 to >1000 folds) of occupational exposure to phthalate residues in various workplaces including PVC and rubber boot/hose manufacturers and nail salons implies the significances of continuous assessment of adverse effects of phthalates and its metabolite (Hines et al., 2009)

The prominent environmental occurrence, significant exposure potential, and the toxicological evidence of dialkyl *ortho*-phthalates ensued the permanent banning of DEHP, DBP, and BBP (>0.1%) in children's toys and articles and an interim prohibition on DOP, DINP, and DIDP (>0.1%) in children's toys in the USA (CPSIA, 2008). Therefore, select dialkyl *ortho*-phthalates are recently being replaced by two major classes of non-phthalate plasticizers – citrates such as ATBC and adiaptes such as DIBA and DEHA in controlled consumer products (USCPSC, 2010; Bernard et al., 2014). ATBC and DEHA are among the U.S. EPA high production volume compounds (>1 M pounds produced or imported). ATBC has been primarily used in cosmetics, food contact wrappings, cables, and children's toys whereas adipates are used in building materials, vinyl floorings, carpet backing, wooden veneer, coated fabrics, and toys (EC, 2008; LCSP, 2011). Although non-phthalate plasticizers are generally considered safe alternatives to phthalate plasticizers, only very few studies focused on evaluating leachability, and toxicological impact of non-phthalate plasticizers in the environment have been reported (Fromme et al., 2016). DEHA and ATBC were previously measured at μg/g levels in PVC medical devices (Gimeno et al., 2014). Recently, Fromme et al. (2016) found DEHA and ATBC in all indoor dust from 63 childcare centers in Germany at a mean concentration of 80 μg/g and 146 μg/g, respectively (Fromme et al., 2016) This was the only study to report non-phthalate plasticizers in indoor dust. In addition, there is no report of phthalate and non-phthalate contamination in indoor dust from salons in the USA