Health risks of residential indoor and outdoor exposure to fine particle-bound phthalates

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ABSTRACT

We performed residential indoor fine particle (PM2.5) measurement from 26 homes and three outdoor monitoring locations. Six PM2.5-bound phthalate easters (PAEs) — including dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (DnBP), butyl benzyl phthalate (BBP), di(2-ethylhexyl) phthalate (DEHP), and di-n-octyl phthalate (DnOP) — were measured using a thermal desorption-gas chromatography/mass spectrometer method. Average concentrations of summation of six PAEs (∑6PAEs) in residential indoors (646.9 ng/m3) were slightly lower than the outdoor levels. DEHP was the most abundant PAE congener (80.3%) and was found at the highest levels, followed by BBP, DnBP, and DnOP. Strong correlations were observed between indoor DEHP with DnBP (rs: 0.88; $p < 0.01$), BBP (rs: 0.83; $p < 0.01$), and DnOP (rs: 0.87; $p < 0.01$). However, no apparent inter-correlations were shown for PAE congeners. Principal component analysis affirmed heterogeneous distribution and notable variations in PAE sources between residential indoor and ambient exposure. The results provide critical information for mitigation strategies, suggesting that PAEs from indoor and outdoor sources should be considered when exploring the inhalation risks of PAEs exposure.

KEYWORDS

Indoor air quality, particle-bound phthalates, DEHP

1 INTRODUCTION

Phthalate esters (PAEs) are a group of synthetic chemicals widely used in polyvinyl chloride (PVC) products and consumer products (such as commodities, medical products, cosmetics, and personal care products) and in households (building materials, furnishing, household goods) (Eichler, Cohen Hubal, & Little, 2019; Meeker, Sathyanarayana, & Swan, 2009). The negative impacts of PAEs on human health have raised global concerns due to their widespread use (IARC Working group, 2000; Katsikantami et al., 2016).

The International Agency for Research on Cancer and the United States Environmental Protection Agency (U.S. EPA) have classified DEHP and BBP as possible human carcinogens (Group B2 and Group C) (Caldwell, 2012; U.S. EPA, 1987). Given that a large proportion (> 85%) of daily time is spent indoors for the general population, research efforts have been made to address PAEs in different indoor microenvironments, including schools, offices, and residential homes (Otake, Yoshinaga, & Yanagisawa, 2004) and from indoor dust (Kang, Man, Cheung, & Wong, 2012). Buildings offer partial protection again ambient origin particulate pollutants, but indoor sources of PAEs enhance the potential for overall exposures. PAEs are physically bound to the plastic polymer and can be easily released into the ambient atmosphere and adhere to indoor particles and settled house dust (Clausen, Liu, Kofoed-Sorensen, Little, & Wolkoff, 2012; Zhang et al., 2021).

The research aims are to (1) examine the occurrence and variations of PM2.5-bound PAE congeners (i.e., DMP, DEP, DnBP, BBP, DEHP, and DOP) in residential indoors; (2) characterize the within- and between- home variability of PAE congeners in residential indoor; (3) investigate the potential sources of PAEs in outdoor and residential indoors.

2 METHODOLOGY

PAE congener concentrations are reported in ng/m3. The Shapiro–Wilk test is used to check the normality of data. Seasonal variations of targeted PAEs were analysed using the Mann–Whitney U test. Differences in PAEs between ambient and residential indoor were calculated using *t* test. The mixed-effects model was used to calculate the within-home variance (σ2w) and between-home variance (σ2b) in residential indoors [37]. Statistical analyses were performed in R 3.5.1. A p-value < 0.05 was considered statistically significant.

Average indoor-to-outdoor (I/O) ratios for PAEs were calculated. We used Spearman's correlation coefficients (rs) to characterize the associations of PAE congeners in and between exposure categories. In addition, we applied principal component analysis (PCA) to identify the potential sources of PAEs in ambient and residential indoor. PCA was performed by using IBM SPSS Statistics (Version 26.0, Armonk, NY, USA: IBM Corp).

3 RESULTS

3.1 Characteristics of PAEs in residential indoor

The reported ∑6PAEs accounted for an average of 1.8 ± 3.7% indoor PM2.5 level (35.1 ± 19.0 μg/m3). Daily residential indoor ∑6PAEs concentrations varied from 0.8 to 3245.4 ng/m3 with an average of 646.9 ng/m3. Average DEHP (582.2 ng/m3) was presented at the highest level in residential indoor PM2.5, followed by BBP (65.5 ng/m3), DnBP (27.1 ng/m3), and DnOP (20.5 ng/m3), accounting for 80.3%, 5.8%, 11.6%, and 1.6% of ∑6PAEs concentrations, respectively. DMP and DEP concentrations were one to two orders of magnitude lower than other PAE congeners because these low-molecular-weight (LMW) PAEs tended to be present in the gas phase.

3.2 Ambient and residential indoor relationships

Comparing the average concentrations of PAEs indoors and outdoors, the average I/O ratios of PAE congeners and ∑6PAEs ranged from 1.8 to 4.8 (Table 1). For paired data, indoor ∑6PAEs exceeded the corresponding outdoor levels in 33.3%–50.0% of households. The median I/O ratios for PAE congeners < 1. As for individual PAE congeners, the highest average I/O ratio was shown for DnBP (4.8), and there were significant differences for outdoor with indoor DnBP exposure ($p = 0.02$), suggesting that DnBP sources are primary in some residential indoors. Further, DMP and DEP concentrations in different exposure categories demonstrated no significant differences but higher average and median I/O ratios compared with other PAE congeners.

3.3 Source identification

Significant correlations were shown between DMP and DEP ($rs = 0.72$, $p < 0.01$) in residential indoor (Table 2). In addition, strong correlations for DEHP with DnBP ($rs = 0.88$; $p < 0.01$), BBP ($rs = 0.83$; $p < 0.01$), and DnOP ($rs = 0.87$; $p < 0.01$) were shown in residential indoor. Similarly, strong correlations were demonstrated outdoors. There are moderate correlations between DnBP with DMP and DEP in residential indoor, suggesting that there might be common sources for these compounds. No such associations were found outdoors. Outdoor monitoring at fixed sites could not capture indoor origin pollutants.

We applied PCA to explore the sources of particle-bound PAEs in PM2.5 (Table 3). For residential indoor PAEs, three principal components accounted for 88.3% of the total variance. Component 1 explained 33.5% of the total variance and comprised BBP (0.90), DnOP (0.93), and a lesser extent of DEHP (0.58), indicating the influence of widely used plasticizers in PVC and other polymer products. Component 2 in residential indoors was loaded with DnBP (0.96)

and DEHP (0.76), which explained 28.0% of the total variance. Component 3 accounted for 26.8% of the data variance and had high loading of DMP and DEP, indicating the household's non-plastic sources (cosmetics, perfumes, and personal care products). The indoor sources of PAEs are more diverse and complicated compared to outdoor sources. It is difficult to disentangle these sources because of a lack of specific observations concerning factors influencing indoor PAE exposure (e.g., plastic products, wall coverings, furniture, and building characteristics).

Table 1. Summary statistics of PAE congeners and ∑6PAEs in residential indoor PM2.5

	Indoor (ng/m^3)							**I/O ratio (no unit)**			
	Mean ± SD	Median	95th	Min – Max	N	σ^2_b (%)	σ^2_w (%)	Mean ± SD	Median	Q1 – Q3[g]	N[d]
DMP	0.17 ± 0.17	0.12	0.43	0.01 – 1.08	61	30.3	69.7	1.9 ± 4.3	0.81	0.52 – 1.46	57
DEP	3.22 ± 3.07	2.21	9.25	0.05 – 16.11	62	23.7	76.3	2.3 ± 5.8	0.84	0.36 – 1.75	60
DnBP	27.1 ± 24.9	21.2	71.10	0.1 – 129.0	63	0	100	4.8 ± 15.2	0.69	0.34 – 1.93	57
BBP	65.5 ± 122.5	12.9	315.6	0.2 – 654.4	53	0	100	1.9 ± 3.5	0.27	0.03 – 1.71	41
DEHP	582.2 ± 604.8	409.5	2007.4	0.4 – 2330.6	59	3.7	96.3	1.8 ± 3.3	0.46	0.16 – 1.75	50
DnOP	20.5 ± 50.1	6.4	98.9	0.1 – 243.0	50	1.6	98.4	2.8 ± 7.5	0.45	0.06 – 3.04	40
$\sum_6$PAEs	646.9 ± 734.1	471.8	2495.5	0.8 – 3245.4	63	7.2	92.8	3.2 ± 11.6	0.46	0.16 – 1.76	55

Table 2. Spearman's correlation matrix for PAEs.

Residential indoor (I)	**DMP**	**DEP**	**DnBP**	**BBP**	**DEHP**	**DnOP**
DMP	1	0.72**	0.30*	-0.003	0.30*	0.05
DEP		1	0.07	-0.16	0.03	-0.04
DnBP			1	0.73**	0.88**	0.76**
BBP				1	0.83**	0.80**
DEHP					1	0.87**
DnOP						1
Outdoor (O)						
DMP	1	0.68**	0.09	0.11	0.11	0.07
DEP		1	-0.0005	-0.16	-0.11	-0.21
DnBP			1	0.77**	0.90**	0.81**
BBP				1	0.90**	0.93**
DEHP					1	0.93**
DnOP						1
I-O	0.14	0.02	0.03	-0.15	-0.13	-0.16
P-I	0.23	0.17	-0.18	0.07	-0.03	-0.02

Table 3. Factor loading of principal component analysis (PCA) on PAEs

	Residential indoor			**Outdoor**	
Species	PC1	PC2	PC3	PC1	PC2
DMP	*	*	0.86	*	0.87
DEP	*	*	0.91	*	0.87
DnBP	*	0.96	*	0.74	*
BBP	0.90	*	*	0.90	*
DEHP	0.58	0.76	*	0.93	*
DnOP	0.93	*	*	0.87	*
Eigenvalue	2.67	1.61	1.00	3.00	1.54
% of variance	33.5	28.0	26.8	50.0	25.7

4 CONCLUSIONS

This investigation revealed a comprehensive picture of the abundance and composition of PAE congeners in outdoor and residential indoors. The within-home variances dominated the total variability of indoor PAE congeners. DEHP was the dominant PAE congener, contributing to 80.3% of ∑6PAEs, followed by BBP, DnBP, and DnOP. The results showed strong

heterogeneity for PAE congeners, and no apparent intercorrelations were observed between outdoor and residential indoors. We further explored the emission sources of exposure to PAEs.

5 ACKNOWLEDGEMENTS

Jiayao Chen acknowledge the TwinAIR project.

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