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Occurrence of Phthalate Diesters in Particulate and Vapor Phases in Indoor Air and Implications for Human Exposure in Albany, New York, USA

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Abstract Phthalate diesters are used as plasticizers in a wide range of consumer products. Because phthalates have been shown in laboratory animal studies to be toxic, human exposure to these chemicals is a matter of concern. Nevertheless, little is known about inhalation exposure to phthalates in the United States. In this study, occurrence of nine phthalates was determined in 60 indoor air samples collected in 2014 in Albany, New York, USA. Airborne particulate and vapor phase samples were collected from various sampling locations by use of a low-volume air sampler. The median concentrations of nine phthalates in air samples collected from homes, offices, laboratories, schools, salons (hair and nail salons), and public places were 732, 143, 170, 371, 2600, and 354 ng/m^3, respectively. Diethyl phthalate (DEP) was found at the highest concentrations, which ranged from 4.83 to 2250 ng/m^3 (median 152) followed by di-*n*-butyl phthalate, which ranged from 4.05 to 1170 ng/m^3 (median 63.3). The median inhalation exposure dose to phthalates was estimated at 0.845, 0.423, 0.203, 0.089, and 0.070 µg/kg-bw/d for infants, toddlers, children, teenagers, and adults, respectively. Inhalation is an important pathway of human exposure to DEP.

Phthalate diesters (or phthalates) are esters of phthalic acid and are used widely as plasticizers in various consumer and industrial products. Phthalates are present in building materials, clothing, personal care products (PCPs), food packaging, toys, vinyl products, lubricating oils, solvents, and detergents (Antian 1973; Hubinger and Havery 2006; United States Environmental Protection Agency [USEPA] 2008; Clausen et al. 2010). Certain cooking utensils, such as spatulas, were reported to contain di(2-ethylhexyl) phthalate (DEHP) and di-*n*-butyl phthalate (DBP) at concentrations of 60–5830 and 60–80 µg/g, respectively (Kawamura et al. 2001). In addition, degassing of DEHP from polyvinyl chloride (PVC) flooring and the emission of DEHP and diisononylphthalate (DiNP) into indoor air from various phthalate-containing products has been reported (Clausen et al. 2012). Diethyl phthalate (DEP) and DBP were found in cosmetics and personal care products at concentration as high as 25,500 and 24,300 µg/g, respectively (Koniecki et al. 2011; Buck Louis et al. 2013; Guo and Kannan 2013; Guo et al. 2014). DEHP was the major phthalate ester found in foods with a median concentration of 28 ng/g in dairy products, 86 ng/g in fish, and 44.5 ng/g in meats from the United States (Schecter et al. 2013). These studies suggest the existence of a wide variety of sources of human exposure to phthalates in the environment.

A few studies have reported the occurrence of phthalates in various indoor environmental samples. A total of 17

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phthalate diesters were found in house dust collected from Canada, and DEHP was found at the highest concentration, ranging from 36 to 3840 µg/g (Kubwabo et al. 2013). The total median concentration of nine phthalates in house dust from China and the United States ranged from 151 to 765 µg/g (Guo and Kannan 2011b). In another study, seven phthalates were measured in house dust from the United States at concentrations that ranged from 1 to 570 µg/g (Bergh et al. 2012).

Although a large number of studies have reported the occurrence of phthalates in house dust, very few have reported the occurrence of these compounds in the airborne particulate and vapor phases of indoor air. DEP (range 145–7120 ng/m^3) and DBP (range 755–14,800 ng/m^3) were reported to occur in indoor air from the United States and Poland (Adibi et al. 2002; Rudel et al. 2003). Fromme et al. (2004) reported the occurrence of DBP in indoor air at median concentrations of 1080 ng/m^3 in apartments and 1190 ng/m^3 in kindergartens in Berlin, Germany. The mean concentrations of six individual phthalates in the indoor air of homes, day care centers, and offices in Stockholm ranged from 4.6 to 1600 ng/m^3 (Bergh et al. 2011). The median concentrations of seven phthalates in indoor air from France were reported at <0.6–326 ng/m^3 (Blanchard et al. 2014). Indoor air is a major source contamination by phthalates in ambient and outdoor air (Cousins et al. 2014). A recent study showed that concentrations of phthalates in indoor air were $\leq$27 times greater than in outdoor air in California (Gaspar et al. 2014). Thus, measurement of phthalates in indoor air will provide an understanding of potential sources and pathways of these chemicals in the environment.

Studies have shown that phthalates elicit reproductive and developmental toxicities in laboratory animals (Gray et al. 2006; Boberg et al. 2008). Specifically, phthalate exposure was shown to be associated with endocrine disruption, respiratory effects, and reproductive and developmental toxicities (Lin et al. 2011; Hauser and Calafat 2005; Calafat and Mckee 2006; Buck Louis et al. 2013). A negative association between environmental phthalate exposure and intelligence or behavior in children has been shown (Cho et al. 2010; Engel et al. 2010). Therefore, if we are to develop strategies to mitigate exposures, a comprehensive assessment of sources of human exposure to phthalates is necessary. Our research group has reported the occurrence of phthalates in foodstuffs, indoor dust, and personal care products in previous studies from the United States (Guo and Kannan 2011b, 2012a, 2013; Guo et al. 2012b, 2014). In the present study, 9 phthalate diesters were determined in 60 indoor air samples collected from Albany, New York, USA. Partitioning of phthalate esters between particulate and vapor phases of indoor air was determined. Furthermore, human exposure to phthalates through the inhalation of indoor air was assessed.

Materials and Methods

Standards and Solvents

Nine phthalate diesters—*i.e.,* dimethyl phthalate (DMP), diethyl phthalate (DEP), diisobutyl phthalate (DIBP), DBP, di-*n*-hexyl phthalate (DNHP), benzyl butyl phthalate (BzBP), dicyclohexyl phthalate (DCHP), DEHP, and di-*n*-octyl phthalate (DOP)—along with their corresponding d_4 (deuterated) internal standards, with a purity of >99 %, were purchased from AccuStandard Inc (New Haven, Connecticut, USA). Analytical-grade acetone was purchased from Macron Chemical (Nashville, Tennessee, USA), and hexane and dichloromethane were purchased from J. T. Baker (Phillipsburg, New Jersey, USA).

Sample Collection and Extraction

Precleaned polyurethane foam (PUF) plugs (ORBO-1000 small PUF; 2.2-cm O.D × 7.6-cm length) were purchased from Supelco (Bellefonte, Pennsylvania, USA). For the analysis of background levels of phthalates, PUFs were extracted with dichloromethane (DCM) and hexane (3:1, v:v) and analyzed by gas chromatography-mass spectrometry (GC-MS). It was found that each of the newly purchased PUF plugs contained DMP, DEP, DBP, DIBP, BzBP, and DEHP at 2.8–5.9, 8.4–46.3, 15.6–70.2, 5.1–33.3, 2.9–10.5, and 21.5–168 ng, respectively ($n = 5$). Therefore, all PUF plugs required additional purification before use. PUFs were purified by shaking with a 100-mL mixture of DCM and hexane (3:1, v:v) for 30 min. This procedure was repeated twice. The cleaned PUFs were wrapped in solvent-rinsed aluminum foil, stored in a glass jar, and placed in an oven at 100 °C until use. The quartz fiber filters (Whatman, grade QM-A, pore size 2.2 µm with a particle retention rating at 98 % efficiency in liquid, 32-mm diameter) were prepared by heating at 450 °C for 20 h. The purified quartz fiber filters were kept in an oven at 100 °C until use. The quartz fiber filters were weighed in an analytical balance (0.01 mg) before and after the collection of air samples for the determination of particle content in air.

Two PUF plugs were packed in tandem in a glass tube (ACE glass Inc., Vineland, New Jersey; 2.2-cm outer diameter × 25-cm length), and the quartz fiber filter was held with a Teflon cartridge (Supelco, PUF filter cartridge assembly) on top of the glass tube packed with PUF plugs. Indoor air samples were collected for 12–24 h by a low-volume air sampler (LP-20; A.P. Buck Inc., Orlando, Forida, USA) at a flow rate of 5 l/min. The total volume of air collected from each location ranged from 3.6 to 7.2 m^3. Air samples (both PUFs and filters) were kept at −18 °C until analysis. The samples were kept frozen for no longer than 3 weeks before analysis. The samples were collected from

January to May 2014 at several locations in Albany, New York, USA. The sampling locations were grouped into six categories: homes ($n = 20$), offices ($n = 7$), laboratories ($n = 13$), schools ($n = 6$), salons ($n = 6$ [hair and nail salons]), and public places ($n = 8$ [e.g., shopping malls]).

Before analysis, samples (both PUFs and filters) were spiked with 100 ng of deuterated internal standards (except for d_4-DEHP, which was spiked at 500 ng). The particulate samples were extracted by shaking glass fiber filters with a mixture of DCM and hexane (3:1; 20 mL; v:v) three times for 5 min each time. The extracts were concentrated in a rotary evaporator at 40 °C to approximately 5 mL. The solution was then transferred into a 12-mL glass tube and concentrated by a gentle stream of nitrogen to exactly 1 mL, which was then transferred into a GC vial.

PUF plugs were extracted by shaking in an orbital shaker (Eberbach Corp., Ann Arbor, Michigan, USA) with DCM and hexane (3:1, v:v) for 30 min. The extraction was performed twice with 100 mL of solvent mixture for the first time and 80 mL for the second time. The extracts were concentrated in a rotary evaporator and then by a gentle stream of nitrogen to exactly 1 mL. The sample was then transferred into a GC vial.

Instrumental Analysis

Nine phthalate diesters were analyzed on a gas chromatograph (6890 N; Agilent, Santa Clara, California, USA) coupled with a 5973 mass spectrometer. A fused-silica capillary column (HP-5MS; Agilent; 5 % diphenyl 95 % dimethylpolysiloxane, 30 m × 0.25-mm inner diameter; 0.5-µm film thickness) was used for the separation of phthalates. Samples were injected in the splitless mode, and the injection volume was 2 µL.

The oven temperature was programmed from 80 °C (held for 1.0 min) to 180 °C at 12 °C/min (held for 1.0 min), increased to 230 °C at 6 °C/min, then to 270 °C at 8 °C/min (held for 2.0 min), and finally increased to 280 °C at 30 °C/min (held for 12.0 min) (Guo et al. 2014). Ion fragments m/z 163, m/z 279, and m/z 149 were monitored for the quantification of DMP, DOP, and seven other phthalate diesters, respectively. The fragment ions m/z 177 for DEP, m/z 233 for DIBP and DBP, m/z 223 and m/z 206 for BzBP, m/z 167 for DCHP, m/z 167 and m/z 279 for DEHP, and m/z 279 for DNHP were monitored for the confirmation of the target compounds (Guo et al. 2012b). Ion fragment m/z 167 was monitored for d4-DMP and m/z 153 for other internal standards.

Quality Assurance and Quality Control

One of the major challenges associated with the analysis of phthalates in air is the potential for contamination from the laboratory materials. Residue levels of phthalates in laboratory materials, including solvents used in extraction, have been studied in our laboratory (Guo and Kannan 2011b, 2012a, 2013; Guo et al. 2011a, 2011c, 2012b, 2014). Before the analysis of air samples, considerable effort was made to decrease the background levels of contamination in the analytical procedures. All glassware was heated at 450 °C for 20 h before use. The baked glassware was covered in clean aluminum foil and kept in an oven at 100 °C until further use. Newly opened solvents were used directly from glass bottles, and exposure of solvent to air was kept minimal. Procedural blanks were analyzed with every batch of samples. Trace levels of DEP (1.9–14.8 ng), DIBP (1.2–11.7 ng), DBP (3.1–22.1 ng), BzBP (1–3.2 ng), and DEHP (3.2–26.1 ng) were found in procedural blanks ($n = 12$) involving new PUFs, and DIBP (0.5–3.3 ng), DBP (1–6.7 ng), and DEHP (2.1–14.9 ng) were found in procedural blanks ($n = 12$) containing quartz fiber filters. All reported concentrations in indoor air samples were subtracted from the mean value found in procedural blanks. The calibration curve was linear over a concentration range from 0.3 to 500 ng/mL for individual phthalate diesters ($R^2 > 0.99$).

A total of 100 ng of internal standards (d_4-phthalates) were spiked into a blank PUF and glass fiber filter (except for d_4-DEHP, which was spiked at 500 ng) and passed through the entire analytical procedure. The average recoveries of internal standards in method blanks were 90–118 % with an RSD that ranged from 5.17 to 11.9 % for PUFs and were 82–116 % with an RSD that ranged from 5.6 to 11 % for the glass fiber filter. The method detection limit (MDL) and the method quantification limit (MQL) were determined based on the lowest point in the calibration standard with signal-to-noise ratios of 3 and 10, respectively; the average volume of air collected, which was 3.6 m^3, and the average mass of airborne particle collected, which was 0.25 mg, were included in the calculation. For the particulate phase, the MQL ranged from 1.5 to 6 µg/g, and for the vapor phase, the MQL ranged from 0.1 to 0.45 ng/m^3 (Supporting Information Table S1). Statistical analysis of the data was performed using Microsoft Excel, Microsoft Office 2010, and Graph Pad Prism, Version 5.0. Concentrations lower than the MQL were assigned a value equal to half the MQL for statistical analysis.

Results and Discussion

Phthalates in Particulate and Vapor Phases in Indoor Air

The mass of airborne particles in air samples was determined based on the difference in the weight of the quartz fiber filter before and after the collection of samples. The mass of particles in air samples ranged from 0.15 to 0.45 mg (mean 0.25). In the particulate phase, DMP, DNHP, DCHP, and DOP were found at a detection frequency of 95, 55, 15, and 15 % respectively (Tables S2 and S3). Nevertheless, DEP, DIBP, DBP, BzBP, and DEHP were found at high concentrations in all of the samples. DEHP, followed by DBP (427 µg/g) and DIBP (370 µg/g), was found at the highest median concentration (465 µg/g) in the particulate phase (Table 1). The total median concentration of sum of nine phthalates in the particulate phase ranged from 1030 µg/g (*i.e.*, approximately 0.1 %) for public places to 14,700 µg/g (*i.e.*, approximately 1.5 %) for salons (hair and nail salons). The overall median concentration of phthalates in airborne particles in 60 samples was 2070 µg/g (approximately 0.2 %). The measured concentrations of phthalate diesters in the particulate phase were similar to those reported for house dust from several countries including the United States and Canada (Bornehag et al. 2005; Guo and Kannan 2011b; Bergh et al. 2012; Kubwabo et al. 2013).

The median concentration of DEP in the vapor phase was 112 ng/m^3, whereas that value in the particulate phase (on a volumetric basis) was 17.3 ng/m^3 (Table S2 and S3). The concentration of DEP was six times greater in the vapor phase than in the particulate phase. Blanchard et al. (2014) reported that the ratio of DEP between the vapor and the particulate phases was 157, which was much greater than the ratios found in our study. Similarly, the DMP concentration in the vapor phase was 33.2 ng/m^3, which was 25 times greater than that in the particulate phase (1.35 ng/m^3). Concentrations of other phthalates (*i.e.*,, DIBP, DBP, BzBP, and DEHP) in the vapor and the particulate phases were not significantly different. DNHP, DCHP, and DOP were found less frequently in indoor air samples (Fig. 1). The median concentration of individual phthalates in the vapor phase ranged from lower than the MQL to 112 ng/m^3, and those in the particulate phase ranged from lower than the MQL to 24.9 ng/m^3.

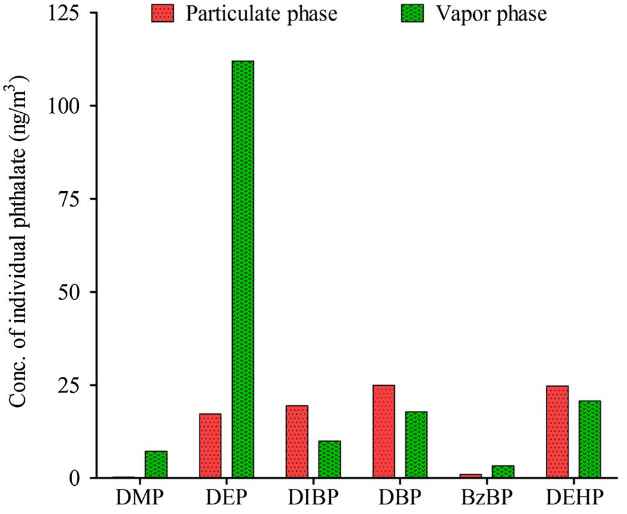

Fig. 1 Median concentrations of individual phthalate esters found in particulate and vapor phases in indoor air from Albany, NY, USA ($n = 60$ samples)

Gas-Particle and Octanol-Air Partition Coefficient of Phthalates

The gas-particle partition coefficient (K_P) and the octanol-air partition coefficient (K_{OA}) of phthalate diesters were calculated on the basis of the concentrations measured in the vapor and particulate phases of indoor air. The partition coefficient, Kp, which has the units of m^3/µg, was determined by Eq. (1):

$$K_P = (F/\mathrm{TSP})/A \tag{1}$$

where F (ng/m^3) and A (ng/m^3) are the particulate and vapor phase concentrations, respectively, and *TSP* (µg/m^3) is the total suspended particulate matter concentration (Finizio et al. 1997; Schossler et al. 2011). F/TSP, which has the unit ng/µg, can be combined to give the fraction of target compound concentration in the particulate phase. Finizio et al. (1997) showed a fundamental relationship between K_{OA} and K_P as shown in Eq. (2):

$$K_P = (f_{om-part} K_{OA})/\rho_{part} \tag{2}$$

By applying $f_{om\text{-}part} = 0.4$ for the organic fraction of dust (Fromme et al. 2005) and a particle density of $\rho_{part} = 1000\ \mathrm{kg/m^3}$ (Turpin et al. 2001; Weschler et al. 2008; Weschler and Nazaroff 2010), Schossler et al. (2011) obtained Eq. (3):

$$\log(K_P) = \log(K_{OA}) - 12.4 \tag{3}$$

We determined K_P and $\log(K_P)$ based on the ratio of concentrations of individual phthalates between the particulate and vapor phases. Equation (3) was used in the calculation of $\log(K_{OA})$ (Table 2). The $\log(K_P)$ and the $\log(K_{OA})$ values of the low molecular-weight phthalates were lower than those of high molecular-weight phthalates. The $\log(K_{OA})$ value ranged from 8.60 for DMP (lowest) to 11.1 for DEHP (highest) (Table 2). In a previous study, the $\log(K_{OA})$ values for six phthalates were reported to range from 6.69 (for DMP) to 12.6 (for DEHP) (Schossler et al. 2011). Nevertheless, our results indicate that the low molecular-weight phthalates, such as DEP and DMP, preferentially partition to the vapor phase, whereas the high molecular-weight phthalates, such as DEHP, tend to partition toward the particulate phase in air.

Concentrations of Phthalates (Particulate Plus Vapor) in Bulk Indoor Air

Total concentrations of individual phthalate diesters in the bulk of indoor air were determined by the summation of

Table 1 Concentrations of phthalate diesters in particulate and vapor phases (ng/m^3) in indoor air from Albany, New York, USA

Building type		Particulate phase						Vapor phase					
		DMP	DEP	DIBP	DBP	BzBP	DEHP	DMP	DEP	DIBP	DBP	BzBP	DEHP
Homes (*n* = 20)	Range	<MQL–2.40	3.42–361	1.47–178	0.85–451	0.11–59.8	4.95–72.0	1.95–83.1	13.2–1630	1.50–80.0	1.09–111	0.20–24.7	2.98–132
	Mean	0.50	79.6	55.3	71.9	6.26	24.1	56.2	463	22.4	21.2	6.22	27.4
	Median	0.27	18.0	33.9	44.7	1.19	22.9	57.5	390	19.6	22.6	2.99	17.4
	DR (%)	100	100	100	100	100	100	100	100	100	100	100	100
Offices (*n* = 7)	Range	<MQL–0.4	0.37–237	1.29–192	3.92–102	0.36–1.95	11.0–52.8	0.41–33.8	4.46–1010	1.64–20.7	1.36–36.4	0.57–17.4	5.49–37.8
	Mean	0.28	42.9	44.3	28.4	0.94	29.7	14.8	248	10.6	18.6	5.97	22.0
	Median	0.25	1.0	7.30	14.3	0.89	29.3	21.5	11.9	9.82	17.0	3.83	10.8
	DR (%)	100	100	100	100	100	100	100	100	100	100	100	100
Laboratories (*n* = 13)	Range	ND–0.36	0.34–14.9	1.48–17.1	2.01–21.3	0.11–4.22	2.48–90.0	0.57–8.48	3.87–28.8	0.85–12.2	1.22–40.7	1.03–20.6	15.4–562
	Mean	0.45	2.57	4.65	6.67	1.40	37.3	4.53	12.5	4.76	8.86	5.81	155
	Median	0.29	1.41	2.43	5.40	0.99	34.0	4.87	10.4	2.62	4.35	3.15	77.0
	DR (%)	46.2	100	100	100	100	100	100	100	100	100	100	100
Schools (*n* = 6)	Range	0.11–0.48	8.89–202	3.64–69.5	9.62–94.3	0.68–8.35	2.04–58.7	6.67–25.9	9.39–280	8.32–67.7	4.41–33.4	0.40–15.9	2.65–72.8
	Mean	0.28	60.3	25.2	44.6	5.45	15.8	13.4	137	30.4	19.7	9.3	18.4
	Median	0.31	40.4	24.1	45.1	7.12	5.93	11.7	134	28.4	20.3	8.6	5.10
	DR (%)	100	100	100	100	100	100	100	100	100	100	100	100
Salons (*n* = 6)	Range	0.25–0.52	17.7–466	12.5–579	35.5–90.8	0.27–1.98	12.4–42.7	23.9–120	897–1940	37.3–802	33.1–1130	0.94–26.0	9.54–663
	Mean	0.39	224	234	65.8	0.88	27.5	96.8	1450	303	473	10.9	195
	Median	0.36	231	225	66.3	0.78	27.5	91.0	1480	151	315	10.7	43.1
	DR (%)	100	100	100	100	100	100	100	100	100	100	100	100
Public places (*n* = 8)	Range	<MQL–1.14	21.6–143	4.71–45.0	11.0–40.2	0.33–3.08	3.89–52.2	1.17–16.2	13.6–675	1.07–104	1.58–203	0.70–17.2	6.66–25.8
	Mean	0.57	54.1	23.2	22.7	1.37	23.0	8.95	246	19.1	68.6	4.42	13.8
	Median	0.49	40.0	24.2	21.4	1.30	23.9	10.9	125	5.68	65.7	3.72	11.8
	DR (%)	100	100	100	100	100	100	100	100	100	100	100	100
Total (*n* = 60)	Range	ND–2.40	0.34–466	1.29–579	0.85–451	0.11–59.8	2.04–90.3	0.41–120	3.87–1940	0.85–802	1.09–1130	0.20–26.0	2.65–663
	Mean	0.35	67.8	53.6	42.8	3.31	27.0	15.5	377	45.7	69.6	5.94	68.3
	Median	1.35	17.3	19.4	24.9	1.02	24.7	33.2	112	19.9	27.8	3.33	20.7
	DR (%)	95	100	100	100	100	100	100	100	100	100	100	100

ND not detectable, *DR* % detection rate, *<MQL* lower than the MQL (range 0.1–0.45 ng/m^3 for individual phthalate)

Table 2 Estimated $\log(K_P)$ and $\log(K_{OA})$ values for phthalate diesters (on the basis of the concentrations measured in particulate and vapor phases in indoor air)

Phthalate diesters	$\log(K_P)$		$\log(K_{OA})$	
	Range	Mean	Range	Mean
DMP	−3.96 to −3.12	−3.80	8.44–9.28	8.60
DEP	−2.99 to −1.83	−2.59	9.41–10.6	9.81
DIBP	−1.75 to −1.45	−1.73	10.5–10.9	10.7
DBP	−2.59 to −1.33	−1.81	9.81–11.1	10.6
BzBP	−2.66 to −1.32	−2.14	9.74–11.1	10.3
DEHP	−1.97 to −1.18	−1.32	10.6–11.2	11.1

$\log(K_P)$ and $\log(K_{OA})$ were estimated based on the concentrations of individual phthalate diesters determined in particulate and vapor phases in indoor air ($n = 60$ samples)

concentrations measured in the particulate and vapor phases and reported on the basis of air volume (m^3). The concentrations of individual phthalate esters determined in bulk indoor air (sum of vapor and particulate phases) are listed in Table 3. DEP was found in all indoor air samples at the highest concentration with values that ranged from 4.83 to 2250 ng/m^3 (median 152). The concentrations of DBP ranged from 4.05 to 1170 ng/m^3 (median 63.3) and DIBP from 2.95 to 1380 ng/m^3 (median 48.8). The measured concentrations of DEP were similar to those reported in indoor air from homes in Stockholm (4.6–1600 ng/m^3) (Bergh et al. 2011) but were six times lower than those reported for indoor air of homes in Krakow, Poland (1000 ng/m^3) (Adibi et al. 2002). A study from Berlin, Germany (Fromme et al. 2004), reported DEP concentrations at 1080 ng/m^3 for apartments and 1190 ng/m^3 for kindergartens, which are within the range of values found in our study.

DNHP, DCHP, and DOP were detected in 41.7, 13.3, and 35 % of indoor air samples, respectively, although their median concentrations were lower than the MDL. Several studies have shown that low molecular-weight phthalate esters (e.g., DEP and DBP) are present in cosmetics and personal care products (Guo et al. 2014). The highest concentration of DEP found in personal care products from the United States was 937 µg/g (approximately 0.9 %, w/w) (Guo et al. 2014). DEP was detected at concentrations ≤38,700 µg/g (approximately 3.9 %), and DBP was found at concentrations ≤59,800 µg/g (approximately 6 %) in cosmetics from Washington, DC, USA (Hubinger et al. 2006). DEP was found in almost all types of surveyed products, and the highest concentrations (25,500 µg/g [2.6 %]) were found in fragrances. DBP was largely present in nail polishes, and a concentration as high as 24,300 µg/g (approximately 2.4 %) was reported from Canada (Koniecki et al. 2011). These results explain high levels of phthalates, especially DEP, found in indoor air in salons (hair and nail salons). The highest measured concentration of DEP in indoor air from salons was 2250 ng/m^3 (median 1680). DIBP and DBP were detected at similar levels in indoor air from salons with a median concentration of approximately 350 ng/m^3. DNHP, DCHP, and DOP were not found in indoor air from salons.

The overall median concentration for the sum of nine phthalates in 60 indoor air samples was 390 ng/m^3. These values are two times lower than those reported from homes in Cape Cod, Massachusetts, USA (1030 ng/m^3) (Rudel et al. 2003). However, our values were similar to the concentrations (450 ng/m^3) reported for residential dwellings in Sapporo, Japan (Kanazawa et al. 2010). Pei et al. (2013) reported 30 times greater levels of five phthalates in indoor air from newly decorated apartments in China (12,000 ng/m^3) than what was found in our study.

A comparison of total concentration of nine phthalates in indoor air among six categories of sampling locations is shown in Fig. 2. Indoor air samples from salons (hair and nail salons) contained the highest total concentration of phthalates (median 2600 ng/m^3), which was one order of magnitude greater than that found in other locations. The concentrations of phthalates measured in other five categories of sampling locations were similar, and the offices had the lowest concentration at 143 ng/m^3.

Composition of Phthalates in Indoor Air

The composition profile of phthalates in indoor air varied among the sampling locations (Fig. 3). Overall DEP, DIBP, DBP, and DEHP, collectively, accounted for ≥94 % of the total phthalate concentrations in indoor air. In homes, schools, salons (hair and nail salons), and public places, DEP was the dominant compound found at 68, 58, 67, and 48 %, respectively, of the total phthalate concentrations. A high proportion of DEP in indoor air was similar to that reported in personal air samples collected in northern Manhattan, New York, USA, which contained DEP at 70 % of the total phthalate concentrations (Adibi et al. 2002). Pei et al. (2013) showed that DEP, BzBP, and DEHP, collectively, accounted for 72 % of the total phthalate concentrations in indoor air from homes in China. Bergh et al. (2011) reported that DEP accounted for >50 % of the total phthalate concentrations in indoor air from Stockholm, Sweden. DIBP and DBP concentrations in indoor air from Albany, New York, USA, were 5–27.2 % of the total phthalate concentrations. DEHP was the dominant compound in indoor air from laboratories (75 %) and offices (42 %). Great proportions of DEHP in laboratories suggest that the sources are predominantly from plastics and PVC products (Rudel and Perovich 2009; Clausen et al. 2010, 2012). The high proportion of DEP and DBP in indoor air can be explained by the fact that these low

Table 3 Concentrations of phthalates (ng/m^3; sum of particulate and vapor phase concentrations) in indoor air from Albany, New York, USA

Building type		DMP	DEP	DIBP	DBP	DNHP	BzBP	DCHP	DEHP	DOP	∑PHT
Homes (*n* = 20)	Range	2.18–85.3	28.0–1780	11.9–253	17.5–472	ND–3.44	0.98–61.3	ND–1.24	11.2–162	ND–1.67	71.9–2820
	Mean	56.7	543	77.8	93.1	<MQL	12.5	<MQL	51.5	<MQL	795
	Median	57.7	432	72.5	90.2	<MQL	3.78	<MQL	45.2	<MQL	732
	DR (%)	100	100	100	100	80	100	10	100	15	–
Offices (*n* = 7)	Range	0.66–34.2	4.83–1250	8.94–209	5.28–138	ND–1.44	1.24–18.2	ND	23.8–81.1	ND–1.76	45.0–1710
	Mean	14.9	291	54.9	47.0	<MQL	6.91	ND	51.6	<MQL	457
	Median	23.5	12.9	18.0	33.5	<MQL	5.77	ND	45.1	<MQL	143
	DR (%)	100	100	100	100	71.4	100	0	100	57.1	–
Laboratories (*n* = 13)	Range	0.57–8.50	6.61–34.6	2.95–27.7	4.05–47.2	ND–2.38	2.09–21.1	ND–1.46	32.5	ND–4.35	49.0–753
	Mean	4.61	15.1	9.42	15.5	0.55	7.19	<MQL	192	0.87	242
	Median	4.09	13.1	6.14	11.9	0.25	4.86	<MQL	111	0.12	170
	DR (%)	100	100	100	100	84.6	100	61.5	100	61.5	–
Schools (*n* = 6)	Range	6.80–26.3	56.1–288	12.1–93.8	21.1–98.7	ND–3.13	2.88–20.5	ND	5.88–92.2	ND–1.36	105–610
	Mean	13.7	197	55.6	64.4	<MQL	11.7	ND	34.2	<MQL	373
	Median	12.1	211	59.0	66.3	<MQL	10.9	ND	10.1	<MQL	371
	DR (%)	100	100	100	100	83.88	100	0	100	50	–
Salons (*n* = 6)	Range	24.4–120	1270–2250	123–1380	122–1170	ND	2.91–27.0	ND	34.4–706	ND	1570
	Mean	97.2	1670	537	539	ND	11.8	ND	222	ND	3050
	Median	91.4	1680	305	373	ND	11.3	ND	77.9	ND	2600
	DR (%)	100	100	100	100	0	100	0	100	0	–
Public places (*n* = 8)	Range	1.34–16.8	35.2–819	9.18–144	22.3–237	ND	1.36–18.8	ND–1.01	16.5–58.8	ND–1.04	86.1–1300
	Mean	9.52	300	42.3	91.3	ND	5.78	<MQL	36.8	<MQL	486
	Median	11.7	156	33.3	81.5	ND	4.45	<MQL	40.6	<MQL	354
	DR (%)	100	100	100	100	0	100	25	100	50	–
Total (*n* = 60)	Range	0.57–120	4.83–2250	2.95–1380	4.05–1170	ND–3.44	0.98–61.3	ND–1.46	5.88–706	ND–4.35	53.6–4850
	Mean	15.9	445	99.3	112	<MQL	9.25	<MQL	85.3	<MQL	778
	Median	34.4	152	48.8	63.3	<MQL	5.64	<MQL	37.9	<MQL	390
	DR (%)	100	100	100	100	55	100	13.3	100	35	–

ND not detectable, *DR* % detection rate, *<MQL* lower than the MQL (range 0.1–0.45 ng/m^3 for individual phthalate), ∑*PHT* total concentrations of nine phthalate diesters

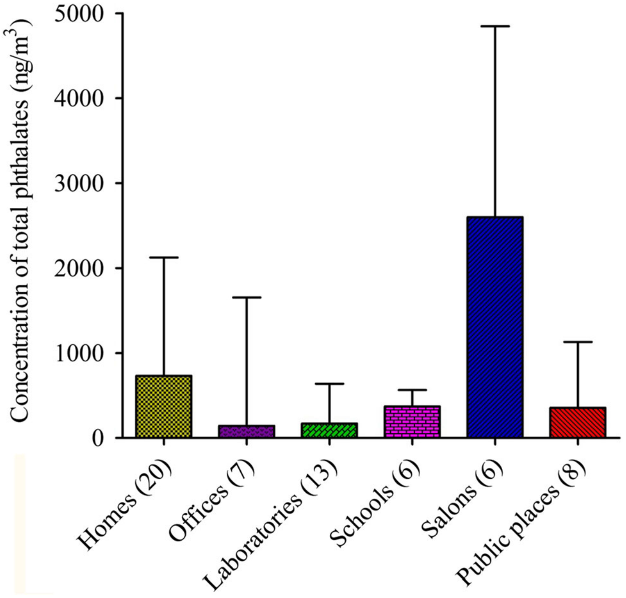

Fig. 2 Total median concentrations with range of phthalate diesters in indoor air from six categories of sampling locations in Albany, New York, USA. Values in *parentheses* refer to the number of samples

molecular-weight phthalates are widely used in cosmetics and personal care products in the indoor environment (Hubinger et al. 2006; Koniecki et al. 2011; Guo and Kannan 2013; Guo et al. 2014).

Human Exposure to Phthalates by Way of Inhalation

Several studies have examined the exposure of humans to phthalates (Koo and Lee 2005; Calafat and MaKee 2006; Clark et al. 2011; Guo and Kannan 2011b, 2013; Guo et al. 2012b, 2014; Schecter et al. 2013). The sources of human exposure to phthalates vary depending on the type of phthalates. For instance, diet is the major source of exposure for DEHP, whereas dermal and inhalation pathways are the major sources of exposure to DEP and DBP (Guo et al. 2014). The contribution of indoor air to phthalate exposure has not been determined previously. We calculated the exposure dose to phthalates through the inhalation of indoor air by multiplying the measured concentrations (μg/m^3) with the volume of air inhaled (m^3). The average air inhalation rate by adults and children was 0.54 m^3/h (13 m^3/d) (CEPA 1994). The estimated median inhalation exposure dose to total phthalates in homes, offices, laboratories, schools, salons (hair and nail salons), and public places were 9.52, 1.86, 2.21, 4.82, 33.8, and 4.60 μg/d, respectively. Among various categories of sampling locations, salons contributed to the highest exposure doses. The overall median value for inhalation exposure to phthalates through indoor air ($n = 60$) was 5.07 μg/d.

The daily inhalation exposure dose of phthalates was calculated for various age groups (Table 4). The calculated daily inhalation exposure doses of total phthalates for infants, toddlers, children, teenagers, and adults were 0.845, 0.423, 0.203, 0.089, and 0.070 μg/kg-bw/d, respectively. These results suggest that phthalate inhalation exposure doses decrease with an increase in age. For DEP, inhalation was the major source of exposure at an exposure dose of 0.027–0.329 μg/kg-bw/d, which was followed by that of DBP (range 0.011–0.137 μg/kg-bw/d), DIBP (range 0.009–0.106 μg/kg-bw/d), and DEHP (range 0.007–0.082 μg/kg-bw/d).

Several earlier studies in our laboratory estimated human exposure to phthalates from various sources in the United States (Guo and Kannan 2011b, 2012a, 2013; Guo et al. 2012b, Guo et al. 2014; Schecter et al. 2013). The contribution of human exposure to phthalates through indoor air inhalation was compared with doses calculated from other exposure pathways (Table 5). The inhalation exposure dose was similar to that calculated through dust ingestion (0.186–1.7 μg/kg-bw/d) (Guo and Kannan

Fig. 3 Composition profiles of six phthalate diesters in indoor air samples from six types of locations in Albany, New York, USA. DNHP, DCHP, and DOP were found less frequently, and their median concentrations were lower than the MQL; therefore, they are not included here

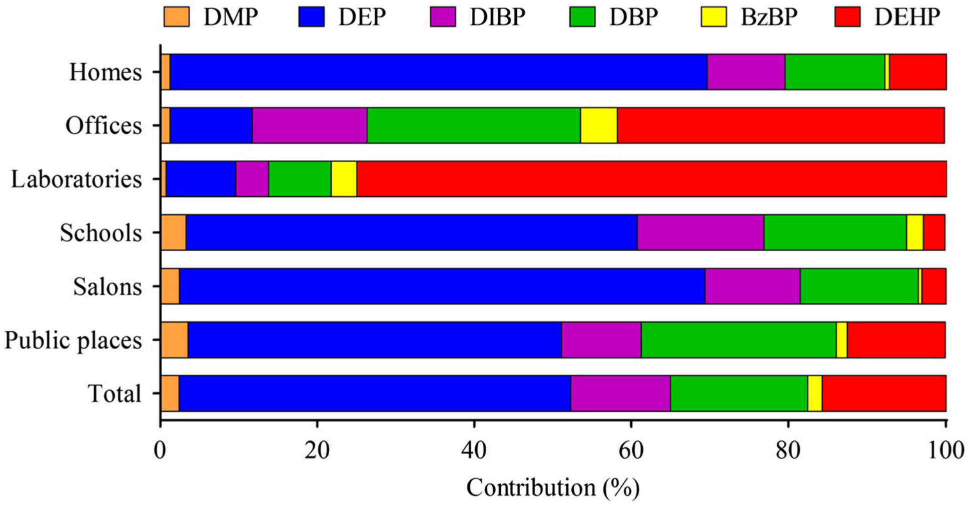

Table 4 Human exposure to individual phthalate diesters through indoor air inhalation in Albany, New York, USA (μg/kg-bw/d)[a]

Age category	DMP	DEP	DIBP	DBP	BzBP	DEHP	∑Exposure
Infants	0.075	0.329	0.106	0.137	0.012	0.082	0.845
Toddlers	0.037	0.165	0.053	0.069	0.006	0.041	0.423
Children	0.018	0.079	0.025	0.033	0.003	0.019	0.203
Teenagers	0.008	0.035	0.011	0.014	0.001	0.009	0.089
Adults	0.006	0.027	0.009	0.011	0.001	0.007	0.070

Infants (<1 y) = 6 kg-bw; toddlers (1–3 y) = 12 kg-bw; children (3–11 y) = 25 kg-bw; teenagers (11–18 y) = 57 kg-bw; adults (>18 y) = 72 kg-bw (Child-Specific Exposure Factors Handbook [USEPA 2008]; ∑Exposure = total daily inhalation exposure dose to nine phthalates by way of indoor air

[a] The average inhalation rate of air for all ages is 13 m^3/d (CEPA 1994)

Table 5 Comparison of human exposure doses to total phthalates through various pathways (μg/kg-bw/d)*

Exposure route	Infants	Toddlers	Children	Teenagers	Adults	References
Dust ingestion	1.21	1.7	0.468	0.291	0.233	Guo and Kannan (2011b)
Dust dermal absorption	0.001	0.0008	0.0006	0.0005	0.0002	Guo and Kannan (2011b)
PCPs (dermal)[a]	0.0095	0.0059	–	–	0.013-0.49	Guo and Kannan (2013)
Diet[b]	–	–	4.68	–	1.03	Schecter et al. (2013)
Indoor air inhalation	0.845	0.423	0.203	0.089	0.070	This study

[a] Exposure dose calculated based on the mean concentration of PCPs (rinse-off, leave-on, and baby care products)

[b] Values are the mean daily dietary intakes of nine phthalates. Food samples (e.g., beverages, milk, fish, fruit, grain, beef, pork, poultry, meat and meat products, vegetable oils, and infant food) were collected from Albany, New York

* USEPA reference doses (RfDs) = 200 μg/kg/d for BBzP (USEPA 2012c), 100 μg/kg/d for DBP (USEPA 2012b), 20 μg/kg/d for DEHP (USEPA 2012c), and 800 μg/kg/d for DEP (USEPA 2012c). The USEPA has not published RfDs for the other phthalates

2011b). The inhalation exposure dose was seven times lower than the exposure dose calculated through dietary exposure (1.03 μg/kg-bw/d for adults and 4.68 μg/kg-bw/d for children) (Schecter et al. 2013). In another study, Guo and Kannan (2013) reported the daily dermal exposure dose, based on the mean phthalate concentrations measured in PCPs from Albany, New York, USA, and the values were 0.0095, 0.0095, and 0.013–0.49 μg/kg-bw/d for infants, toddlers, and adult females, respectively. Accordingly, the daily exposure dosage of total phthalates from PCPs was approximately 100 times lower than the inhalation exposure dose. However, it should be noted the indoor air is an important contributor to DEP exposure. The exposure dose calculated for individual phthalates through various pathways was lower than the currently published USEPA reference doses (USEPA 2012a, 2012a, 2012a, 2012a).

Conclusions

Concentrations of nine phthalate diesters were determined in 60 indoor air samples from homes, offices, laboratories, schools, salons (hair and nail salons), and public places (shopping malls) in Albany, New York, USA, in 2014. Median concentrations of total phthalates in indoor air ranged from 143 to 2600 ng/m^3, and the highest levels were found in hair salons. DEP accounted for 40 % of the total concentrations in indoor air. Inhalation exposure to phthalates ranged from 0.070 to 0.845 μg/kg-bw/d, and inhalation is a major source of exposure to DEP. The current level of phthalate exposure in the United States is lower than the USEPA's reference doses. Studies have reported emission of phthalates from vinyl flooring and crib mattress covers in homes (Liang and Xu 2014, 2015). The increase in the use of such products in buildings would increase the environmental emission and human exposure to these compounds. This study establishes baseline levels for future environmental assessment of phthalates.

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