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Pollution levels and characteristics of phthalate esters in indoor air of offices

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ABSTRACT

The pollution status and characteristics of PAEs (phthalate esters) were investigated in indoor air of offices, and PAEs of both gas-phase and particulate-phase were detected in all the samples. The concentration (sum of the gas phase and the particulate phase) was 4748.24 ng/m^3, ranging between 3070.09 and 6700.14 ng/m^3. Diethyl phthalate, dibutyl phthalate, and di(2-ethylhexyl) phthalate were the most abundant compounds, together accounting for 70% of the ∑6PAEs. Dividing the particulate-phase PAEs into four size ranges (<2.5, 2.5–5, 5–10, >10 μm), the result indicated that PAEs in $PM_{2.5}$ were the most abundant, with the proportion of 72.64%. In addition, the PAE concentration in $PM_{2.5}$ correlated significantly with the total particulate-phase PAEs ($R^2 = 0.85$). Thus, the amount of PAEs in $PM_{2.5}$ can be estimated from the total amount of particulate-phase PAEs using this proportion. In a comparison between the offices and a newly decorated study room, it was found that pollution characteristics were similar between these two places. Thus, it is implied that the PAE concentration decreased by 50% 2 yr after decorating.

Introduction

Plasticizers are important promoters in material processing, and are widely used in building materials, packaging materials, electronics, medical parts and many other necessities of life (Zhang et al., 2012; Wang et al., 2010a, 2010b; Schripp et al., 2010; Afshari et al., 2004; Clausen et al., 1999; Sheldon et al., 1993; Pöhner et al., 1997). As the most widely used plasticizer, phthalate esters (PAEs) are ubiquitous in indoor environments, and have become emerging organic pollutants in indoor air (Bornehag et al., 2005; Koch et al., 2003; Wensing et al., 2005).

PAEs are considered to be endocrine-disrupting compounds that have functions similar to female sex hormones. These compounds can destroy testicles, inhibit the formation of sperm, affect reproductive functions, and damage the reproductive system of males (Hauser et al., 2006; Li et al., 2006; Swan et al., 2005; Silva et al., 2005; Foster et al., 2001). In addition, PAEs also have influences on females. They can cause precocious female puberty (Adibi et al., 2003; Colón et al., 2000) and increase the risk of breast cancer (Swan and Davis, 2003). What's worse, reports (Pei et al., 2013; Wang et al., 2010a, 2010b) indicated that PAEs may pose carcinogen risks to human health.

The respiratory system, digestive tract, and skin are the main pathways for PAEs entering the body; and concentrations of PAEs have been detected in human urine, blood, milk and many other bodily fluids (Hines et al., 2009a, 2009b; Mortensen et al., 2005). Compared with food and water, PAE exposure in the air is more common. So far, research on PAE pollution in indoor air has mainly focused on either the gas phase (Wensing et al., 2005) or particle phase (Wang et al., 2012; Lin et al., 2009; Kolarik et al.,

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Table 1 – Concentration of PAEs in indoor air of offices (ng/m^3). G: gas-phase; P: particulate-phase.

Site	DMP		DEP		DBP		BBP		DEHP		DnOP		∑PAEs		Total
	G	P	G	P	G	P	G	P	G	P	G	P	G	P	
1	320.08	180.04	597.07	332.07	711.65	340.18	571.97	299.62	720.66	690.75	nd	nd	2933.43	1842.67	4776.09
2	364.48	233.17	550.18	371.59	690.52	366.06	429.68	202.65	620.55	642.67	nd	nd	2655.37	1816.15	4471.52
3	566.06	311.62	858.66	559.41	1011.21	848.07	589.62	395.77	811.38	748.35	nd	nd	3836.92	2863.22	6700.14
4	510.65	245.97	708.68	417.18	606.15	414.66	263.66	189.97	564.52	504.89	nd	nd	2723.59	1846.35	4569.94
5	820.62	490.65	790.58	445.29	613.11	569.07	309.40	233.11	684.52	593.67	nd	nd	3218.22	2331.79	5550.01
6	598.12	310.66	805.98	417.65	617.56	478.43	287.65	239.45	536.99	467.78	nd	nd	2846.31	1913.97	4760.27
7	432.27	210.72	488.40	385.02	775.10	380.58	408.22	210.01	566.47	505.73	nd	nd	2670.45	1692.06	4362.51
8	663.73	374.88	746.92	389.68	675.74	452.62	379.32	204.24	611.46	507.87	nd	nd	3077.18	1929.28	5006.46
9	496.42	244.92	653.05	361.75	463.80	294.32	544.53	234.75	506.89	416.98	nd	nd	2662.70	1552.72	4215.41
10	537.73	230.75	380.06	168.28	422.96	260.31	342.13	173.92	290.78	263.16	nd	nd	1973.67	1096.43	3070.09
mean	532.22	283.33	657.96	384.79	658.78	440.42	419.61	245.72	591.22	534.18	nd	nd	2859.78	1888.46	4748.24

DEP: diethyl phthalate, DBP: dibutyl phthalate, BBP: butylbenzyl phthalate, DEHP: di(2-ethylhexyl) phthalate; DnOP: di-n-octyl phthalate; PAE: phthalate esters.

2008; Bornehag et al., 2005), but very few studies have investigated both gas-phase and particle-phase PAEs simultaneously in the indoor environment. Offices are important places for people to live and work; many people spend 8 hr a day or more in offices, which contain a lot of packaging materials and electronic parts. As a result, PAEs may be present at high levels in offices. However, there have been few studies on PAE pollution in offices, and studies on particle size distribution are even fewer. In this study, 10 offices decorated for over 2 yr were selected as sample sites, and the concentration level and pollution characteristics of PAEs (gas-phase and particulate) and size distribution of particulate PAEs in offices were investigated.

1. Materials and methods

1.1. Chemicals and materials

All chemicals and solvents used for extraction and gas chromatography (GC) analysis were of HPLC grade. Phthalate standard mixtures M-8060, including dimethyl phthalate (DMP), diethyl phthalate (DEP), dibutyl phthalate (DBP), butylbenzyl phthalate (BBP), di(2-ethylhexyl) phthalate (DEHP) and di-n-octyl phthalate(DnOP), were purchased from AccuStandard (New Haven, CT, USA) as stock solutions in isooctane. The concentration of each phthalate was 2.0 mg/mL.

1.2. Sample analysis

1.2.1. Sample collection

Indoor air samples were repeatedly collected from 10 offices in Hangzhou, from the spring to the fall in 2013. Before sampling, all the doors and windows were kept closed for 24 hr, and during our experiment, all the doors and the windows were kept closed, while normal residential activities were maintained in the offices. Sampling collections were divided into 2 parts.

In the 1st part, both gas-phase and particulate-phase PAEs were simultaneous determined, using a scientific and well-developed SVOC sampling method (Pei et al., 2013; Zhu et al., 1997): the samples were collected into a plexiglass sampling head with a glass fiber filter (37 mm in diameter, pore size of 0.45 μm, Whatman, England) and subsequently into a glass tube packed with 2 g XAD-2 adsorbent (Sigma-Aldrich, USA), using an electronically controlled air sampler (PC-A, Hengda, Zhejiang, China). The sampling device was set 1.5 m above the floor, with sampling times of 8 to 10 hr, and sampling flow of 1.0 L/min (air pump changes were in the range of less than 5% before and after sampling).

The 2nd part was particulate classification sampling. There are 4 sampling heads on the sampler, including a $PM_{2.5}$ sampling head, PM_5 sampling head, PM_{10} sampling head and TSP sampling head. When sampling $PM_{2.5}$, all the sampling heads were used successively; when sampling PM_5, the $PM_{2.5}$ sampling head was taken out; other fractions were collected similarly. The samples were collected with a glass fiber filter (90 mm in diameter, pore size of 0.45 μm, Whatman, England) using a medium volume sampler (Hengda, Zhejiang, China). The sampling device was set 1.5 m above the floor, with sampling times of 1–2 hr, and sampling flow of 60 L/min (air pump changes were in the range of less than 5% before and after sampling).

When sampling was finished, both ends of the sampling device were kept under seal, and samples were brought back to the laboratory for immediate processing. Temperature, humidity, and air pressure were synchronously recorded using an electronic temperature and humidity instrument (HTC-1, Boyang, Zhengzhou, China) and a digital air pressure monitor (BY-2003P, Taishi, Suzhou, China), respectively.

1.2.2. Sample pretreatment and analysis

Samples were extracted from XAD-2 and glass fibers filter using an ultrasonic cleaner (SK250HP, KUDOS, Shanghai, China) for 30 and 25 min, respectively, with a mixture of dichloromethane/acetone (1:1) as the extraction solvent. Then, half of the extraction liquid with 30 μL dimethyl sulfoxide added was condensed using a N-EVAP (MTN-2800W, Automatic Science, Tianjin, China). The sample was analyzed by a gas chromatograph (FULI 9790, Wenling, China) with a DB-5 capillary column (30 m × 0.25 mm ID × 0.25 μm, Agilent Technology Inc, America). The analysis was performed using the pulsed split-less mode, and the injection volume was 2 μL. High purity nitrogen was used as the carrier gas. The column condition was 60°C, held for 2 min, increased to 240°C with a rate of 15°C/min, followed by a rate of 5°C/min to 280°C, held for 10 min.