



Characterization and flux assessment of airborne phthalates released from polyvinyl chloride consumer goods

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ARTICLE INFO

Keywords:

Phthalates
Polyvinyl chloride
Thermal desorption
Gas chromatography/mass spectrometry
Flux

ABSTRACT

The concentrations and fluxes of airborne phthalates were measured from five types of polyvinyl chloride (PVC) consumer products (vinyl flooring, wallcovering, child's toy, yoga mat, and edge protector) using a small chamber (impinger) system. Airborne phthalates released from each of those PVC samples were collected using sorbent (Tenax TA) tubes at three temperature control intervals (0, 3, and 6 h) under varying temperature conditions (25, 40, and 90 °C). A total of 11 phthalate compounds were quantified in the five PVC products examined in this study. To facilitate the comparison of phthalate emissions among PVC samples, their flux values were defined for total phthalates by summing the average fluxes of all 11 phthalates generated during the control period of 6 h. The highest flux values were seen in the edge protector sample at all temperatures (0.40 (25 °C), 9.65 (40 °C), and 75.7 $\mu\text{g m}^{-2} \text{h}^{-1}$ (90 °C)) of which emission was dominated by dibutyl isophthalate. In contrast, the lowest fluxes were found in wallcovering (0.01 (25 °C) and 0.05 $\mu\text{g m}^{-2} \text{h}^{-1}$ (40 °C)) and child's toy (0.23 $\mu\text{g m}^{-2} \text{h}^{-1}$ (90 °C)) at each temperature level. The information regarding phthalate composition and emission patterns varied dynamically with type of PVC sample, controlled temperature, and duration of control.

1. Introduction

The environmental significance of semi-volatile organic compounds (SVOCs) has garnered a great deal of attention over the past years due to their potential impacts on indoor air quality (Jarnstrom et al., 2009). Increased levels of SVOCs have been observed to affect human health through various diseases such as allergies, respiratory disease, and nervous system dysfunction (Marc et al., 2012). The major components of SVOCs in indoor air have generally been explained by such chemicals as phthalates (used as plasticizers), organophosphate-based flame retardants, and phosphororganic compounds (Wensing et al., 2005). Most of these chemicals are produced by and released from a wide range of sources including building materials, consumer products, and office equipment (Destailats et al., 2008; Liang and Xu, 2015).

Since high-molecular weight compounds have relatively high boiling points (or low vapor pressures), phthalates are recognized as one of the infamous pollutants present in various environmental media including indoor air and surface dust (Clausen et al., 2004). In addition, resuspension of phthalates from sedimented dust has also been regarded as a major source of low indoor air quality (Clausen et al., 2004). As reported in many recent studies, exposure to phthalates and their metabolites may exert a number of adverse effects on human

health such as irreversible changes in the development of the reproductive organs (Liang et al., 2015). In fact, a relationship between exposure to phthalates (at the normal concentrations in indoor) and allergic symptoms in children has also been demonstrated (Hulin et al., 2012). It was also suggested that phthalate exposure is a potential contributor to a disorder of neurodevelopment and/or obesity (Bu et al., 2016).

Phthalates can be released from various indoor sources such as wall coverings, wall paints, vinyl flooring, furniture, and electronic devices (Wensing et al., 2005). In most studies of phthalates emissions, the focus has been on a limited number of phthalate targets like di(2-ethylhexyl) phthalate (DEHP) due to their relative abundance in indoor environments and related health concerns (Cao et al., 2017; Clausen et al., 2004; Noguchi and Yamasaki, 2016; Wu et al., 2016; Xu and Little, 2006). The United States Environmental Protection Agency (U.S. EPA) has started to implement practice guidelines for the production and emission of eight phthalates including diisobutyl phthalate (DIBP), di-*n*-octyl phthalate (DnOP), diisononyl phthalate (DINP), and diisodecyl phthalate (DIDP). These compounds are being considered due to their widespread use in consumer products and the possibility of pervasive human/environmental exposure, as described in a recent report (U.S. EPA, 2012a). Accordingly, DINP had the highest production

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Table 1A
Basic information for 11 (seven main targets plus four references) phthalate compounds investigated in this study and the relevant quality assurance (QA) results. (A) Seven target phthalates quantified by external calibration analysis.

Order	Compound	Short name	Formula	CAS number	Molecular weight (g.mol ⁻¹)	Boiling point (°C)	Mass to charge value ^a (m.z ⁻¹)	Retention time (min)	RSD ^b (%)	MDL ^c (ng m ⁻³) ^c		Flux MDL (ng m ⁻² h ⁻¹) ^f				
										(ng m ⁻³) ^c	(ng m ⁻³) ^c	VF	WC	CT	YM	EP
1	Dimethyl phthalate	DMP	C ₁₀ H ₁₀ O ₄	131-11-3	194	284	163	13.0	0.66	0.03	6.78	1.08	0.31	4.52	0.87	1.62
2	Diethyl Phthalate	DEP	C ₁₂ H ₁₄ O ₄	84-66-2	222	295	149	13.9	1.92	0.04	10.2	1.63	0.47	6.83	1.31	2.45
3	Dibutyl phthalate	DBP	C ₁₆ H ₂₂ O ₄	84-74-2	278	340	149	16.0	1.76	0.11	26.7	4.24	1.21	17.8	3.42	6.37
4	Benzyl butyl phthalate	BBP	C ₁₉ H ₂₀ O ₄	85-68-7	312	370	149	18.1	0.27	0.03	7.50	1.19	0.34	5.00	0.96	1.79
5	Di(2-ethylhexyl) adipate	DEHA	C ₂₂ H ₄₂ O ₄	103-23-1	371	417	129	18.2	2.39	0.01	3.42	0.54	0.16	2.28	0.44	0.82
6	Di(2-ethylhexyl) phthalate	DEHP	C ₂₄ H ₃₈ O ₄	117-81-7	391	385	149	19.2	0.87	0.35	88.1	14.0	4.00	58.7	11.3	21.0
7	Di- <i>n</i> -octyl phthalate	DnOP	C ₂₄ H ₃₈ O ₄	117-84-0	391	380	149	20.9	3.19	0.02	4.54	0.72	0.21	3.03	0.58	1.08

^a Major mass spectrum was used for the quantitation of each compound.

^b Relative standard deviation (determined using liquid working standard of 10 ng μL⁻¹).

^c Method detection limit.

^d Method detection limit value in terms of absolute mass amount.

^e Method detection limit value in terms of mass per volume assuming 4 L volume of air.

^f Method detection limit value equivalent to flux values for each PVC sample.

volume (about 500 million pounds), followed by DIDP and DEHP (U.S. EPA, 2006). Although DEHP recorded the highest production volume (about 152 million pounds) in 2012 among those phthalates, a large amount of DINP and DIDP were also produced such as over 108 and 0.5 million pounds, respectively (U.S. EPA, 2012b). In addition, moderate toxic effects of benzyl butyl phthalate (BBP), DIBP, DINP, and DIDP were verified in aquatic organisms (e.g., algae and freshwater fish) (Staples et al., 1997). Although investigation on the emission rates of diverse phthalates (e.g., di-*n*-butyl phthalate, benzyl butyl phthalate, diisononyl phthalate, and di(2-ethylhexyl) adipate) in the indoor environments has recently increased (Bi et al., 2015; Liang and Xu, 2014), it is still highly desirable to acquire reliable data on the concentration levels and emission rates of various phthalates for the accurate assessment of their exposure routes or potential effects on human health (Ejaredar et al., 2015).

The purpose of this study was to understand the emission characteristics of diverse phthalate compounds (n = 11) produced from five types of plastic/rubber products under three temperature conditions. It was noted that temperature has been considered as one of the most significant parameters to describe emission behavior of phthalates in recent studies (Bi et al., 2015; Cao et al., 2017; Liang and Xu, 2014). The quantification of 11 phthalates was made based on two different approaches: (1) the external calibration method for seven target compounds by liquid standards and (2) prediction based on the effective carbon number (ECN) method for four reference compounds without standards (Kim and Kim, 2013; Szulejko et al., 2013; Szulejko and Kim, 2014). For the latter species lacking suitable liquid (or gaseous) standards, concentration data were estimated using the ECN method. In this manner, emission properties and flux values of 11 phthalates were compared in relation to (1) type of plastic material, (2) temperature, and (3) duration of temperature control for the emission of target samples. Unlike many previous studies, this research offers valuable insights into emission behavior of diverse phthalates in various respects by expanding quantifiable species of phthalates into unprecedentedly large scale (e.g., up to 11 species). Hence, based on their concentration data from different types of plastic materials, emission behavior of total phthalate (or the most abundant species) is assessed in both absolute and relative sense over varying temperature conditions.

2. Materials and methods

Emission concentrations and fluxes of airborne phthalates generated from five different types of polyvinyl chloride (PVC) products (vinyl flooring, wallcovering, child's toy, yoga mat, and edge protector) were investigated using a dynamic flux chamber (impinger) system. To characterize emission behaviors of different phthalates under varying conditions, samples were collected using sorbent tubes (ST) at three temperature control intervals (0, 3, and 6 h; elapsed time from the point when the temperature reached 25, 40, or 90 °C) by controlling temperatures at three different levels (25, 40, and 90 °C). Hence, 45 data points (5 × 3 × 3) were obtained for each phthalate species investigated in this study. Initially, the emission of phthalates was measured under room temperature conditions (25 °C). For comparative purposes, analysis was extended to two other temperatures of 40 °C to simulate human body temperature and 90 °C to investigate the changes in phthalate emissions at the uppermost temperature condition acquired by the heating system (temperature controller). This maximum temperature condition was added mainly for research purposes although it can be observed only under some special conditions in indoor environment (e.g., inside automobile under very hot weather conditions). Phthalate samples adsorbed onto the sorbent tubes were then analyzed using a combination of thermal desorption (TD) and a gas chromatography (GC)/mass spectrometry (MS) system. Sampling, pre-treatment, and analytical procedure used in this study was referred to Occupational Safety & Health Administration (OSHA) method and some previous studies (Aragon et al., 2013; Liang and Xu, 2015; OSHA,

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