



## Characteristics of Phthalates in the Indoor Air of Computer Classrooms

FENG-HSIANG CHANG and CHUN-FU CHIU\*

Department of Leisure, Recreation and Tourism Management, Tzu Hui Institute of Technology, Pingtung 926, Taiwan

\*Corresponding author: Fax: +886 8 8647123; Tel: +886 8 8647367, Ext.157; E-mail: max1006@hotmail.com; chang.taiwan@gmail.com

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We evaluated the airborne phthalate concentrations inside and outside of two computer classrooms and one office located on a Taiwanese college campus. The four phthalates with the highest median concentrations in indoor air were diisobutyl phthalate (DIBP, 2012.5 ng/m<sup>3</sup>), di-*n*-butyl phthalate (DBP, 329.4 ng/m<sup>3</sup>), di-2-ethylhexyl phthalate (DEHP, 167.6 ng/m<sup>3</sup>) and diethyl phthalate (DEP, 149.6 ng/m<sup>3</sup>). Only DBP was detected in the outdoor air, with a median concentration of 174.3 ng/m<sup>3</sup>. The reference outdoor DBP concentrations were approximately 3-100 times lower than the present indoor DBP concentration results. This strongly indicates that the emission sources were present in the indoor environment. Furthermore, when personal computers were turned on, the concentrations of DEP, DBP, DIBP and DEHP in the indoor air increased and DEHP was a note worthy characteristic phthalate. Opening the doors and windows immediately after entering a computer classroom and decreasing the indoor particle concentrations are both suggested to reduce the exposure of users to DEHP.

**Keywords:** Phthalates, Indoor, Computer classroom, Diisobutyl phthalate, Di-2-ethylhexyl phthalate.

### INTRODUCTION

Phthalates are commonly used as plasticizers in polyvinyl chloride (PVC) plastics. Because phthalate plasticizers are not chemically bound to PVC, they can leach, migrate, or evaporate into the atmosphere, foodstuff, or other materials<sup>1</sup>. This process accelerates as plastic products age and break down<sup>2</sup>. In the past decade, the sources, potential health effects and measured air concentrations of phthalates have been comprehensively reviewed and documented<sup>1,3,4</sup>.

Phthalates are present in a wide variety of products including vinyl upholstery, shower curtains, food containers and wrappers, toys, floor tiles, lubricants, sealers, adhesives, paints, capacitor dielectrics, medical equipment, drugs and cosmetics<sup>1,2,4-7</sup>. Furthermore, personal computers and electronic devices, which are used extensively, may also contain phthalates<sup>8-12</sup>. The uses of various phthalates partially depend on the molecular weight (MW) of the phthalates<sup>4</sup>. High-MW di-2-ethylhexyl phthalate (DEHP), diisononyl phthalate (DiNP) and diisodecyl phthalate (DiDP) are the phthalates produced in the highest volume for use in construction materials, clothing and furnishings. These phthalates are primarily used to impart flexibility to PVC plastic. Low-MW phthalates such as diethyl phthalate (DEP), dimethyl phthalate (DMP) and di-*n*-butyl phthalate (DBP) are typically used as solvents and in adhesives, waxes, inks, cosmetics, insecticides and pharmaceuticals. Single applications may also use mixtures of phthalates.

Risk assessments of phthalates have been performed by the US Agency for Toxic Substances and Disease Registry (ATSDR) and the International Agency for Research on Cancer (IARC)<sup>1</sup>. Several phthalates have been reported to interfere with androgen production in animal studies, with developing male fetuses being the most sensitive to this effect<sup>3</sup>. Animal-based studies have demonstrated that after DEHP treatment, mature female rats exhibited decreased blood estradiol levels, a prolonged estrous cycle and inhibited ovulation<sup>13</sup>. A metabolite of DBP, monobutyl phthalate (MBP), was also reported to affect the reproductive function of the uterus in female rats<sup>14</sup>. Furthermore, cell culture experiments revealed that the major metabolite of DEHP (MEHP) causes decreased aromatase expression in rat ovarian granulosa cells<sup>15</sup>. However, the toxic effects of phthalates on humans has not been fully established yet and remain controversial<sup>16,17</sup>.

Nevertheless, it is reported that an association between the maternal levels of urinary phthalate metabolites and reproductive tract development in male offspring in the general population<sup>18</sup>. An association between sperm quality and urinary phthalate metabolite levels in adult men was also reported<sup>19</sup>. Epidemiological studies have revealed that women with endometriosis exhibited higher DEHP concentrations in their plasma than did healthy women<sup>20</sup>. Girls whose blood concentrations of phthalates were higher than those of the control group exhibited early-onset breast development, indicating that phthalates can affect the normal development of female

physiology<sup>21</sup>. Children and adolescents may be particularly susceptible to the toxic effects of ethylhexyl phthalate esters on their physiological and reproductive development<sup>22</sup>. Therefore, although animal studies suggest that current exposure levels among the general population are below levels of health concern, several studies on humans with typical levels of exposure have demonstrated that adverse health effects caused by phthalates can still occur.

Phthalate metabolites have been detected in nearly all of the human urine samples tested in previous studies, indicating widespread exposure<sup>1,23</sup>. Bergh *et al.*<sup>24</sup> surveyed indoor air concentrations of phthalates in 169 apartments and reported that the measured phthalate concentrations in the air ranged up to 11000 ng/m<sup>3</sup>, with PVC flooring being a major source of benzyl butyl phthalate (BBP) in indoor air. Otake<sup>25</sup> collected indoor air from 27 family homes in the Tokyo metropolitan area and reported that DBP exhibited the highest average concentration (0.75 ± 1.17 µg/m<sup>3</sup>), followed by DEHP (0.32 ± 0.60 µg/m<sup>3</sup>) and BBP (0.02 ± 0.03 µg/m<sup>3</sup>). However, these DBP and DEHP median concentrations were lower than those reported by Fromme<sup>5</sup>, who analyzed the air in 74 nursery rooms in Berlin (DBP, 1.18 µg/m<sup>3</sup>; DEHP, 0.458 µg/m<sup>3</sup>). Bergh *et al.*<sup>26</sup> conducted a survey of 30 sampling sites representing three indoor environments (private homes, day-care centers and workplaces) in Stockholm, to obtain representative concentration profiles of phthalates in ambient air. Their analytical results revealed that DEP (420-3900 ng/m<sup>3</sup>) was the most abundant analyte in all of the air samples, followed by DBP (190-2300 ng/m<sup>3</sup>). Samples from workplaces and day-care centers had similar concentration profiles.

Inhalation exposure has typically been considered relatively minor<sup>1</sup>. However, correlations have been observed between the levels of DEP, DBP and BBP urinary metabolites and indoor or personal air concentrations of the parent phthalate, suggesting that indoor air is a primary route of exposure or is a proxy for another route, namely dermal absorption<sup>27, 28</sup>.

Technical and vocational college students now spend more time using computers and stay longer in computer classrooms than they did in the past. Our previous studies have demonstrated that insufficient ventilation rates and accumulated contaminants in computer classrooms, as well as the emission of phthalic carbamates from computer equipment, may lead to health risks<sup>29,30</sup>. Therefore, the long-term exposure of students

and instructors to these contaminants is worthy of further attention and research. However, few studies have considered this problem and there is still a lack of regulations on phthalate concentrations in indoor air in Taiwan.

The major aims of this study were: (i) to investigate the concentration profiles of phthalates in various indoor and outdoor environments (ii) to compare the phthalates levels between indoor and outdoor air and (iii) to elucidate the possible sources of the phthalates present in indoor air.

## EXPERIMENTAL

The chemicals used in this study were purchased from Accu Standard Inc. (New Haven, CT, USA) and Chem Service Inc. (West Chester, PA, USA), and are listed in Table-1. The chemicals were dissolved in toluene to prepare stock standard solutions.

A gas chromatograph/mass spectrometer (GC/MS) was used for the analysis of phthalates. The instrumental and analytical conditions are listed in Table-2.

TABLE-2  
INSTRUMENTAL AND ANALYTICAL CONDITIONS

Item	GC/MS
GC model	Agilent Technologies 6890N Network GC system
Column	Agilent DB-5MS; 30 m × 0.25 mm × 0.25 µm
Autosampler	Agilent 7683 Automatic Liquid Sampler
Injection mode	pulsed splitless
Injector temperature	250 °C
Carrier gas type	99.999 % helium
Carrier gas mode	Constant flow at 1 mL/min
Program of GC oven	Initial temperature at 70 °C, 30 °C/min to 155 °C; 10 °C/min to 235 °C, hold for 2 min; 10°C/min to 280 °C, hold for 1 min
MS model	Agilent Technologies 5973 Network Mass Selective Detector
MS mode	SIM mode

The methods for sampling and analyzing phthalates in air were modified from Otake *et al.*,<sup>31</sup> and only brief descriptions are given here.

**Sampling:** The phthalates in indoor and outdoor air were sampled by passing the air through a 7 cm × 4 mm glass tube containing two layers of charcoal granules (SKC Tested and

TABLE-1  
CHEMICALS USED IN THIS STUDY

Compound name	Abbreviation	m.w.	Provider
Dimethyl phthalate	DMP	194.18	Accu Standard
Dimethyl phthalate (ring-d <sub>4</sub> )	DMP-d <sub>4</sub>		Chem Service
Diethyl phthalate	DEP	222.24	Accu Standard
Diethyl phthalate (ring-d <sub>4</sub> )	DEP-d <sub>4</sub>		Chem Service
Di- <i>n</i> -butyl phthalate	DBP	278.34	Accu Standard
Di- <i>n</i> -butyl phthalate (ring-d <sub>4</sub> )	DBP-d <sub>4</sub>		Chem Service
Diisobutyl phthalate	DIBP	278.34	Chem Service
Benzyl butyl phthalate	BBP	312.36	Accu Standard
Benzyl butyl phthalate (ring-d <sub>4</sub> )	BBP-d <sub>4</sub>		Chem Service
Dicyclohexyl phthalate	DCHP	330.42	Chem Service
Di-2-ethylhexyl phthalate	DEHP	390.56	Accu Standard
<i>Bis</i> (2-ethylhexyl)phthalate (ring-d <sub>4</sub> )	DEHP-d <sub>4</sub>		Chem Service
Di- <i>n</i> -octyl phthalate	D <sub>n</sub> OP	390.56	Accu Standard
Di- <i>n</i> -octyl phthalate (ring-d <sub>4</sub> )	D <sub>n</sub> OP-d <sub>4</sub>		Chem Service

Certified Sorbent Sample Tubes; 20/40 mesh; Chrom 102, Cat. No. 226-107; Eighty Four, PA), namely a sampling layer and a breakthrough layer, which contained 100 mg and 50 mg of charcoal, respectively. The air flow rate was 1000 mL/min and each sampling took 3 days. Thus, approximately 4.3 m<sup>3</sup> of air was passed through the charcoal tube, using an SKC Air Check 2000 pump.

Indoor air samples were obtained from two computer classrooms (four samples from classroom A and two samples from classroom B) and an office (two samples) on a college campus located in the rural area of Pingtung, Taiwan, when the doors and windows were kept closed. All of the indoor sampling environments contained no decorations and only the office had eight chairs containing polyurethane foam (PUF). The sampling devices were placed on a desktop located in the center of the indoor area. Depending on the research requirements, electrical appliances including computers, monitors and lights were either switched on or off during sampling. Samplings were performed between July 9 and July 20, 2010. The indoor and outdoor air samples were collected simultaneously, and a total of four outdoor air samples were collected.

**Analysis of indoor air:** Phthalates adsorbed in the charcoal tube were extracted into 1 mL of toluene using ultrasonication at 20 kHz for 10 min. The ultrasonic bath that was used was a SONICS VCX 750 (Sonic & Materials Inc., Newtown, CT, USA). After extraction, the toluene was separated from the charcoal using centrifugation (3000 rpm × 5 min). The analytical method that was used is summarized as follows: The limits of quantitation detection (LQD) for all the phthalates were 10 pg. Limit of quantitation detection is defined as the construction of a calibration curve in which the standard solution with the lowest concentration, containing the minimal absolute amount of the analyte, was injected. The precision of the analyses, represented as relative standard deviations (RSDs) at 0.1 µg/mL, was less than 20 % for all of the phthalates (0.57-14.03 %). The mean recoveries of the phthalates from the charcoal were greater than 80 % (81.06-95.68 %). The quality control tests included in the air sample analyses comprised a break through test, quality control sample analysis, blank sample analysis, duplicate sample analysis and spiked sample analysis.

## RESULTS AND DISCUSSION

**Quality assurance:** For all of the samples, the values obtained by analyzing the breakthrough layer were lower than

the LQD, indicating that no break through phenomena occurred. Analysis of the quality control sample (250 ng/mL) revealed a tolerance error between -7.6 % (DCHP) and 10.4 % (D<sub>n</sub>OP), which was acceptable because it was between -20 and 20 %; thus, the calibration curve was applied. Each batch of samples collected over 3 consecutive days was subjected to a blank sample analysis and a duplicate sample analysis. The concentrations of the phthalates detected in the blank samples were all below the LQD. The RSDs of the duplicate sample analyses were all less than 20 %. A spiked sample analysis was performed on every 10 samples; this involved the recovery of phthalates (500 ng/mL), which had been sampled from 4.3 m<sup>3</sup> of indoor air prior to spiking, spiked into charcoal tubes. The recoveries of the phthalates from coexisting matrices of indoor air were within the range of 80-120 % for all of the compounds except DIBP, which exceeded the linear range).

**Phthalate concentrations in indoor and outdoor air:** Table-3 shows the means, standard deviations (SDs), median values and minimal and maximal concentrations of phthalates in the indoor and outdoor air of the sampling sites under investigation. Among the phthalates analyzed, DIBP, DBP, DEHP, and DEP were frequently detected indoors and at higher concentrations than those of the other compounds; the median concentrations of these phthalates were 2012.5, 329.4, 167.6 and 149.6 ng/m<sup>3</sup>, respectively. BBP, DCHP and D<sub>n</sub>OP were not detected in the indoor air samples. DMP was detected in only one indoor air sample. Table-4 demonstrates that the amount of phthalates in the indoor air were almost the same as that reported in previous relevant studies<sup>5,24-26,32,33</sup>. The DBP concentration is nearly one of the highest in the relevant literature. DCHP and D<sub>n</sub>OP were not detected or not measured, and an elevated DMP concentration was detected exclusively in study of Fromme *et al.*<sup>5</sup>. The phthalate with the highest concentration in air samples seems to vary according to country. DBP was reported to be the most abundant phthalate in studies conducted in Germany and Japan<sup>5,25,33</sup>, whereas in studies from the United States and Sweden<sup>26,32</sup>, DEP was the most abundant. We detected the highest concentration of DIBP ever reported, which was up to four times higher than that reported in other studies. DIBP is an additive used to keep plastics soft or flexible (plasticizer) and is frequently used in applications such as nitro cellulose plastic, nail polish, explosive materials and lacquer manufacturing, often in combination with other phthalates. Presently DIBP has not been evaluated at the EU level and is

TABLE-3  
MEANS, STANDARD DEVIATIONS (SDs), MEDIAN AND MINIMUM AND MAXIMUM VALUES OF THE MEASURED CONCENTRATIONS OF PHTHALATES IN THE INDOOR AND THE OUTDOOR AIR OF THE SAMPLING SITES

Compound	Indoor air sample (n = 8)				Outdoor air sample (n = 4)			
	Mean ± SD (ng/m <sup>3</sup> )	Median (ng/m <sup>3</sup> )	Minimum (ng/m <sup>3</sup> )	Maximum (ng/m <sup>3</sup> )	Mean±SD (ng/m <sup>3</sup> )	Median (ng/m <sup>3</sup> )	Minimum (ng/m <sup>3</sup> )	Maximum (ng/m <sup>3</sup> )
DMP	3.4 ± 6.3	< 2.3	< 2.3	18.9	< 2.3 ± 0.0	< 2.3	< 2.3	< 2.3
DEP	206.1 ± 165.3	149.6	20.7	497.6	< 2.3 ± 0.0	< 2.3	< 2.3	< 2.3
DBP	332.6 ± 171.9	329.4	64.4	588.3	171.8 ± 20.1	174.3	147.2	191.3
DIBP	1840.5 ± 951.0	2012.5	630.1	3092.6	4.9 ± 7.4	< 2.3	< 2.3	16.00
BBP	< 2.3 ± 0.0	< 2.3	< 2.3	< 2.3	< 2.3 ± 0.0	< 2.3	< 2.3	< 2.3
DCHP	< 2.3 ± 0.0	< 2.3	< 2.3	< 2.3	< 2.3 ± 0.0	< 2.3	< 2.3	< 2.3
DEHP	136.6 ± 87.7	167.6	< 2.3	250.9	< 2.3 ± 0.0	< 2.3	< 2.3	< 2.3
D <sub>n</sub> OP	< 2.3 ± 0.0	< 2.3	< 2.3	< 2.3	< 2.3 ± 0.0	< 2.3	< 2.3	< 2.3

Mean values are calculated by using half values of limit of quantitation detections in case of being below limit of quantitation detections.

TABLE-4  
MEDIAN AIR CONCENTRATIONS OF PHTHALATES ( $\text{ng}/\text{m}^3$ ) IN RESIDENCES AS REPORTED IN RELEVANT STUDIES

Country	Year	Location type	n	DMP	DEP	DBP	DIBP	BBP	DCHP	DEHP	D <sub>10</sub> OP
Germany <sup>5</sup>	2004	Apartments	59	436	643	1083	459	18	NA	156	NA
Germany <sup>5</sup>	2004	Kindergarten	74	331	353	1188	505	NA	NA	458	NA
Japan <sup>25</sup>	2004	Homes	27	NA	100	390	NA	10	70	110	NA
USA <sup>32</sup>	2010	Homes	40	NA	330	140	130	6.8	NA	68	NA
Japan <sup>33</sup>	2010	Houses	40	48	61	200	75	NA	NA	147	NA
Sweden <sup>24</sup>	2011	Apartments	169	16	210	190	230	8.9	NA	220	NA
Sweden <sup>26</sup>	2011	Homes	10	15	1300	850	270	21	NA	200	NA
Sweden <sup>26</sup>	2011	Day care centers	10	4.7	870	600	190	21	NA	240	NA
Sweden <sup>26</sup>	2011	Work places	10	4.4	620	550	230	15	NA	100	NA
This study		Computer class rooms and offices	8	NA	149.6	329.4	2012.5	NA	NA	167.6	NA

NA means values not reported or below reported determination limit

not subject to any European Union ban. Because the application properties of DIBP are similar to those of DBP (the use of which was banned in the manufacturing of toys, childcare articles, and cosmetics), it can be used as a substitute for DBP in PVC, paints, printing inks and adhesives<sup>34</sup>. Whether DIBP was detected at elevated concentrations in this study because it was used as a substitute for DBP is unclear.

Phthalates are subject to photodegradation, biodegradation and anaerobic degradation and, thus, generally do not persist in the outdoor environment<sup>35,36</sup>. Table-3 shows that the median concentration of DBP in outdoor air was  $174.3 \text{ ng}/\text{m}^3$  and the concentrations of all the other phthalates were below the LQD. An elevated outdoor DBP concentration was observed in this study compared with those measured in Sweden ( $1.7 \text{ ng}/\text{m}^3$ )<sup>37</sup>, Japan ( $22 \text{ ng}/\text{m}^3$ )<sup>38</sup>, Germany ( $3.1 \text{ ng}/\text{m}^3$ )<sup>39</sup>, France ( $19 \text{ ng}/\text{m}^3$ )<sup>40</sup>,

the Netherlands ( $10 \text{ ng}/\text{m}^3$ )<sup>41</sup> and China ( $59 \text{ ng}/\text{m}^3$ )<sup>42</sup>. The reference outdoor DBP concentrations were approximately 3-100 times lower than those measured indoors in the present study. This strongly indicated that the emission sources of DBP were present in the indoor environment, which is consistent with the assumption of Otake *et al.*<sup>25</sup>.

**Effects of computer use on indoor concentrations of phthalates:** Previous studies have indicated that personal computers and electronic devices may be possible sources of indoor phthalates<sup>8-12</sup>. In the computer classrooms surveyed, the analytical results revealed that the concentrations of airborne DEP, DBP, DIBP and DEHP increased when 50 sets of personal computers (PCs) were switched on (statistically insignificant; Fig.1). The lowest detected concentration increased as the number of PCs in use increased, particularly

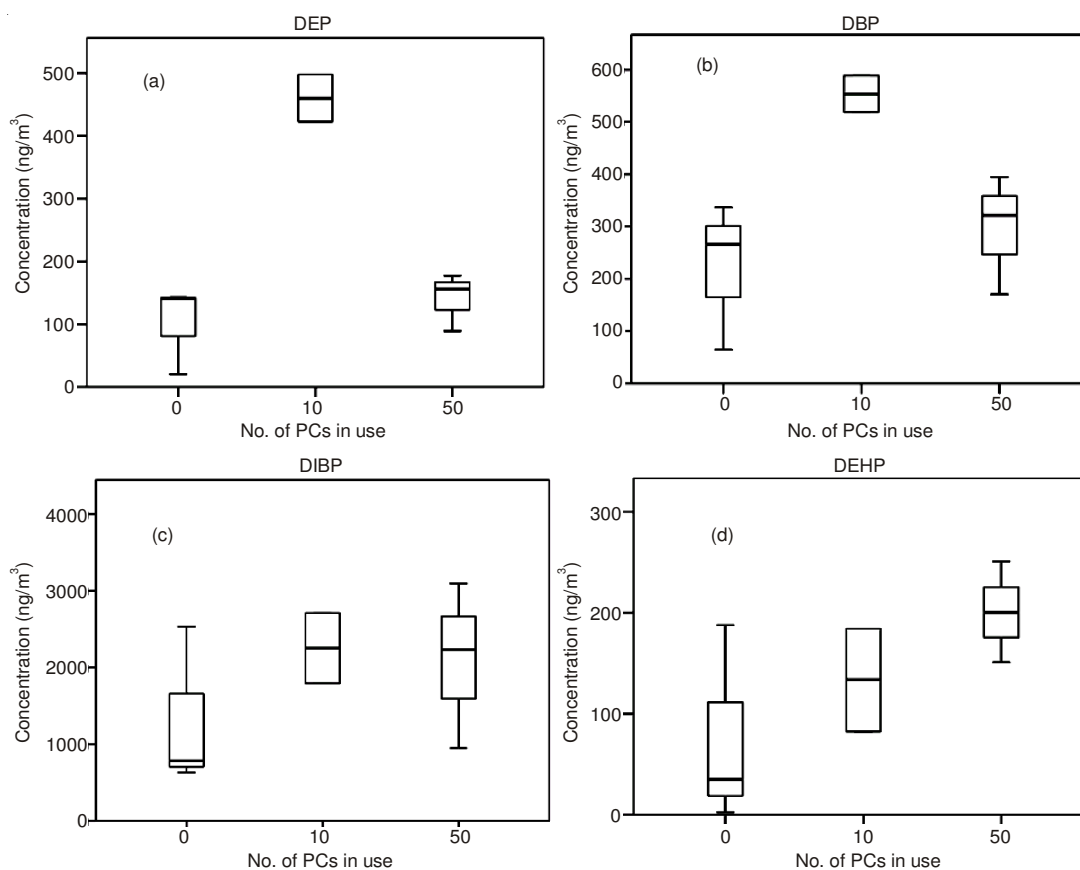


Fig. 1. (a)(b)(c) In computer classroom, when all computers are switched on (50 PCs in use), the lowest concentrations of DEP, DBP, and DIBP are higher than not boot (0 PCs in use), but not statistically significant at the  $\alpha = 0.05$  level. (d) The airborne concentration of DEHP increases with the number of PCs in use within indoor environments, but not statistically significant at the  $\alpha = 0.05$  level

for DEHP, but the median values were not significantly different (Fig. 1d). When the PCs were turned on, we observed an increase in the lowest concentrations and in the median values of the compounds. The data indicated that PCs being operated likely released DEP, DBP, DIBP and DEHP into the indoor air. Furthermore, DEHP may be a characteristic phthalate emitted by PCs. This result is consistent with that of Hikwama<sup>9</sup>.

## Conclusion

In this study, DIBP was the most abundant phthalate in indoor air and its concentration was higher than that of other phthalates. Based on current knowledge, this finding has not been reported previously. Whether this is the result of laws restricting the types of phthalates being used in other countries or because the use of certain phthalates has been reduced is yet to be confirmed. Furthermore, this study revealed that the presence and use of computer equipment may increase the indoor phthalate concentration. When the PCs were turned on, the concentrations of DEP, DBP, DIBP and DEHP in the indoor air increased and DEHP was a note worthy characteristic phthalate. Benning *et al.*<sup>43</sup> reported that the emission rate of DEHP from vinyl flooring was substantially enhanced in the presence of particles. Xu *et al.*<sup>44</sup> tested the emission rate of phthalates from vinyl flooring in a custom-designed chamber and reported that the DEHP concentration increased slowly and reached steady state after approximately 40 days. Based on the findings of Benning *et al.*<sup>43</sup> and Xu *et al.*<sup>44</sup> regarding the emission of DEHP from vinyl flooring, we suggest decreasing the concentration of particulates within computer classrooms to reduce the emission rate of DEHP from PCs. In addition, opening windows to provide fresh air can help reduce the levels of inhalable exposure to phthalates.

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