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Letter

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Phthalate Esters on Hands of Office Workers: Estimating Influence of Touching Surfaces

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ABSTRACT

Phthalate esters are known to be transferred to hands by contact with surfaces, however, little is known about associations between masses on hand wipes and frequency or duration of touching surfaces, especially surfaces in office environments. Relationships between PAEs on hands and multiple surfaces in offices were investigated. Wipes of hands, computers, and mobile phones as well as dust on furniture were collected from 55 offices in China. Positive associations were found between masses of di-2-ethylhexyl phthalate (DEHP), dibutyl phthalate (DnBP), benzyl butyl phthalate (BBP) and di-*n*-octyl phthalate (DnOP) on wipes of hands and wipes of keyboards of computers. When workers used keyboards with polymer covers (dust covers), masses of these lipophilic PAEs on hands were significantly correlated with masses on keyboards rather than dust on furniture. For workers who used keyboards without polymer covers, masses on hands were related to masses in dust on furniture. Use of polymer covers containing PAEs and less washing of hands could increase the mass of exposure via hand to body of office workers, which could further result in as much as 10-fold greater hazard. Thus, more hand washing and less use of polymer products containing PAEs were recommended for office workers to reduce exposures.

INTRODUCTION

Phthalate esters (PAEs) are used as plasticizers to enhance flexibility in a variety of household and building materials, including cables, wires, silicone films, screen guards, vinyl flooring and polyvinylchloride (PVC) tubing [1]. Phthalate esters including di-2-ethylhexyl phthalate (DEHP), dibutyl phthalate (DnBP), dimethyl phthalate (DMP), diethyl phthalate (DEP), benzyl butyl phthalate (BBP) and di-*n*-octyl phthalate (DnOP) (Table S1)[2, 3] are not chemically bound within products and can be slowly emitted into indoor environments. In a study that analyzed 89 organic chemicals in 120 homes, phthalate esters were one of the most abundant indoor pollutants [4].

Recently, indoor exposure of humans to PAEs on skin and especially on hands has been shown to be non-negligible [5, 6]. Phthalate esters that are transferred to hands by contact with surfaces and direct air-to-skin transport, can represent a significant proportion of total accumulation [6]. Besides direct absorption through skin, PAEs on hands can result in inadvertent ingestion, by biting of nails, sucking of digits or eating finger foods [7, 8].

Since previous studies of PAEs have focused on residential exposures, little is known about exposure in offices, although studies have shown greater concentrations of PAEs in dust from offices than those from apartments [9]. Phthalate esters which tended to be strongly adsorbed to indoor surfaces, such as protectors of computer screens, keyboards, furniture, floor and desks, are commonly touched by hands of office workers [10]. Whether and how touching surfaces in offices can increase penetration through hands and further increase the health risk was unknown.

It was hypothesized that PAEs in office furniture might result in exposure of office workers through touching surfaces. The main objectives of the present study were to examine relationships between PAEs on hands of people and on probable touched surfaces in offices by determining amounts of PAEs on surfaces in offices, including computers, mobile phones and dust on furniture. Another objective was to better understand the role of behaviors of workers, including use of polymer covers and habits of hand washing to these relationships and also to the hazards of exposure via hand to body.

MATERIALS AND METHODS

Collection of samples. Samples were collected from April to June 2014. Participants for this study consisted of 55 adults who worked at least 20 h a week in an office and were healthy, nonsmokers in the cities of Huaian (n=12), Zhenjiang (n=31) and Nanjing (n=12) in East China. Offices were located in ten different buildings, and only one person per office participated in the study. Questionnaires were also completed at the time samples were collected (see study design in SI). Within-individual variability was estimated by collecting samples in triplicate from four participants. Samples of dust were collected from each participant's office, including dust on desks, chairs, bookcases and personal accessible furniture by use of a standardized protocol that used a mini vacuum cleaner.

To avoid contamination of hands from sources other than the offices, participants were asked to wash their hands before entering the office. Samples of PAEs on hands were collected by use of wipes applied at close of business. A 7.6×7.6 cm sterile gauze pad pre-cleaned with dichloromethane and acetone was immersed in 3.0 mL of isopropyl alcohol (Tedia Co. Ltd, Fairfield, OH, USA) and then used to wipe the palm and back of the hand from wrist to fingertips, including sides. Wipes of computer keyboards and mobile phones were collected from participants in their office after collection of wipe samples of hands and at least 1 day after the last time the computer, phone or furniture was cleaned.

To confirm sources of PAEs, wipes of keyboards were further collected from twelve computers in six offices. In each office, there were two keyboards, one with a polymer cover and one without a polymer cover. Polymer keyboard covers are removable polymer plastic sheets that are placed on keyboards and left in place while typing. Keyboards and polymer covers were cleaned with isopropyl alcohol in the morning and they were kept in offices without disruption for 5 days. More information for collection of samples is given in supporting information (SI).

Preparation of samples and quantification of PAEs. PAEs in samples were extracted by use of accelerated solvent extraction (ASE). PAEs were quantified by use of a Thermo TSQ Quantum Discovery triple-quadrupole mass spectrometer (San Jose, CA, USA) in multiple-reaction monitoring (MRM) mode. More detailed information for instrumental analysis, procedural blanks and other QA/QC procedures is listed in SI, Table S1 and Table S2.

Assessment of cumulative hazard. The concept of relative cumulative hazard quotient (HQ), developed previously, was employed to incorporate cumulative exposure into the assessment [11]. The daily intake (DI) of PAEs through exposure on hands was divided by tolerable daily intake values for each PAE (TDI_i value) and the HQ for individual PAEs ($HQ\text{-}PAE_i$) were summed to obtain total hazard quotients (HQs) including BBP, DBP, DEP and DEHP.

$$\text{HQs} = \sum_{i=1}^{n} \text{HQ-PAE}_i = \sum_{i=1}^{n} \frac{\text{DI}_i(\mu\text{g/kg bw/day})}{\text{TDI}_i(\mu\text{g/kg bw/day})} \qquad (1)$$

Where: DIi is daily intake (exposure through hand) of individual PAEs, and n is the number of PAEs considered. DIi is the sum of exposure through hand-to-mouth contact (μg/kg bm/day) and absorption through the dermis of the hand (μg/kg bm/day) (More detail is shown in SI and Table S3).

Data analysis. We used Spearman correlations (SPSS statistics software package, V. 17.0) to determine associations between continuous variables while minimizing the influence of outliers. To minimize effects of skewed data and outliers, we created categorical variables from surfaces and hand wipe data: Two-level variables (lesser, and greater) were created using the median as a cut-point (detail is in SI).

RESULTS AND DISCUSSION

Phthalate esters on wipes of hands. Phthalate esters were detected in all wipes of surfaces of hands (Table S4). As expected, DEHP was the predominant compound, followed by DnBP and DnOP. This pattern for skin wipes is consistent with the pattern observed in serum of people from China for DEHP, DnBP and DnOP, although no study has previously reported masses of DnOP on wipes of hands [12, 13]. Few studies have examined phthalates in wipes of hands (Table S5). Generally, DEHP and DnBP are among the most widely detectable PAEs on wipes of hands in

Korea, Beijing in China and also East China detected in the present study [14, 15]. DEP was not detected on wipes of hands of Koreans, while a frequency of detection of less than 20% was observed in the study of people in Beijing, which is approximately the same as the frequency of 52% observed in the present study. This might be due to its greater vapor pressure, which results in a greater partition coefficient [16].

Phthalate esters from surfaces in offices. PAEs were detectable on surfaces of furniture, keyboards and phones, which based on the results of questionnaires, were the most touched surfaces (Table S4). DnBP, BBP, DEHP and DnOP had a greater detection frequency for all types of surface samples. DEHP was the most predominant compound, with geometric means (GM) of 148 μg/g, 83.6 μg and 10.2 μg for dust, keyboards and phones, respectively. For all PAEs, masses on keyboards were 3- to 8-fold greater than those on phones, which might be due to the large area of keyboards and limited variation indicated similar sources for PAEs on keyboard and phone. Distributions of masses and normalized concentrations of PAEs measured on wipe samples had similar variability (Figure S1), and masses not concentrations on wipes of hands, surfaces of computer keyboards and phones were used in the following studies. Concentrations and masses of PAEs in repeated samplings of dust and wipes over a three month period are mostly consistent (See repeated measurements in SI and Figure S2).

Associations of PAEs on hand wipes with indoor surfaces. Total masses of PAEs on wipes of hands were positively correlated with masses on wipes of surfaces of computer keyboards, with a Spearman correlation coefficient (R) of 0.59 (p <0.001, Figure S3). Among the six PAEs, masses of DEHP and DnBP on wipes of keywords and hands were most correlated with Spearman correlation coefficient (R) of 0.58 (p <0.001) and 0.47 (p <0.001), respectively (See Figure S4 and S5). Results of linear regression showed that DEHP and DnBP on surfaces of computer keyboards were a significant predictor of PAEs on hands (p < 0.001, Table S6). Masses of DEP and DMP on wipes of computer keyboards were not associated with masses on hands. This indicated that behavior of congeners with shorter alkyl side chains, such as DEP and DMP, in the indoor environment might differ from that of longer chain congeners[17]. Results of previous studies have indicated that shorter-chain PAEs are more hydrophilic, thus they might be less likely to stick to surfaces of hands, especially for longer periods of time[18].

Total masses of PAEs on wipes of hands were weakly correlated with concentrations in office dust and wipes of surfaces of phones. Similar results were obtained for other individual phthalates. Previous studies have also shown weak associations between masses of penta-BDE, BDE-183 or BDE-209 in office dust with masses on hands. [19, 20, 21].

Predictors of masses of PAEs in wipes of hands. Masses of total PAEs were further categorized according to the number of times hands were washed during work. Participants who washed their hands fewer than four times per day had on average about 10-fold greater masses of Σphthalate esters on their hands compared with those with a greater frequency of washing. Similar situations were observed for masses of DnBP, BBP, DEHP and DnOP on less frequently washed hands which were 3-, 7-, 16- and 7-fold greater than hands which were washed more frequently (Table S6).

According to the questionnaire, participants who washed their hands less than four times per day were further divided into two categories according to wheter they used a polymer keyboard covers or not(Table S6). Masses of the sum of PAEs on wipes of hands of those who used polymer covers (n=24) were approximately 14-fold greater than masses on hands of individuals who did not use polymer covers on keyboards (n=15), although concentrations in dust from offices for the two categories were similar. Similar trends were observed for DEHP and DnBP on wipes of hands, which were 4- and 18-fold greater, respectively. Masses of DEHP and DnBP on polymer keyboard covers were significantly correlated with masses on hands of those who used polymer covers ($r=0.88$, $p<0.001$ for DEHP and $r=0.66$, $p<0.001$ for DnBP), and the correlations were stronger than those for the person who did not use polymer covers on keyboards ($R=0.57$, $p<0.001$ for DEHP and $R=0.38$, $p<0.001$ for DnBP). For persons who used polymer covers, the greater masses of PAEs on their hands were likely caused by direct contact with polymer covers during working hours.

Participants in offices with lesser concentrations of ΣPAEs, DnBP, BBP, DEHP and DnOP (with ΣPAEs less than median masses) in dust who used polymer covers (n=10) had significantly (p<0.01) greater masses of PAEs on wipes of their hands compared with people who worked in offices with greater concentrations (with ΣPAEs greater than median masses) in dust, but did not use polymer covers (n=14, Figure 1). For those who did not use polymer covers, participants in offices with greater concentrations of PAEs in dust had significantly (p<0.01) greater masses of PAEs on wipes of their hands. This result indicated that for workers who did not use polymer covers on their keyboards, redistributions of PAEs in office environments influenced masses on hands and concentrations in dust on furniture were correlated with masses on hands. Mass on hands of workers who used polymer covers on their keyboards were less related to concentrations in office dust, but more related to masses on polymer covers, which seems to be the source of the PAEs on hands.

Influence to hazards of exposure via hand to body. Estimated HQs of total exposure via hands during working was 0.03 (50^{th} centile of masses) and 0.8 (95^{th} centile of masses), which were less than 1.0, but by a small margin of safety (1.0 / 0.8 =1.3) (Table 1). The mean cumulative HQ for participants who washed their hands less than four times was twice the amount for those who washed more than four times per day. Among the participants who washed their hands less than four times per day, the HQs for people who used keyboard polymer cover was 0.18, and is as much as 10-fold greater than that for people who do not use polymer keyboard covers.

Among multiple surfaces in offices, exposure to PAEs through hands are likely affected by exposure to PAEs on surfaces of computer keyboards, especially when keyboard polymer covers are used. Washing of hands and not using of polymer keyboard covers could reduce the mass and related hazard of exposures of office workers to PAEs, via hands. Nowadays, polymer products including polymer keyboard covers and protective films for phone/pad screens that contain PAEs are widely used and act as the most common touchable surface according to our questionnaire survey with total contact being as much as 10 hours per day. These polymer products contain PAE as supplements or impurities. Significant migration of PAE from some polymer packaging of foods or oil have been investigated previously. However, to our knowledge, little information is available from which to evaluate migration of PAE from polymer surfaces to hands. Since direct contact with these may be an underappreciated source of exposure of humans, more attention should be paid to the additive compositions of polymer products in general.

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ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website.

Supporting experimental section, results section with detailed description, Tables S1−S6, and Figures S1−S5.

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Figure Legends

Figure 1. Geometric means (GM) of masses of phthalate esters on wipes of hands by amount of dust in office and polymer cover using categories ($n = 39$). Greater masses were found in lesser-dust/used category than greater-dust/unused category ($p < 0.05$).

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306 Figure 1.

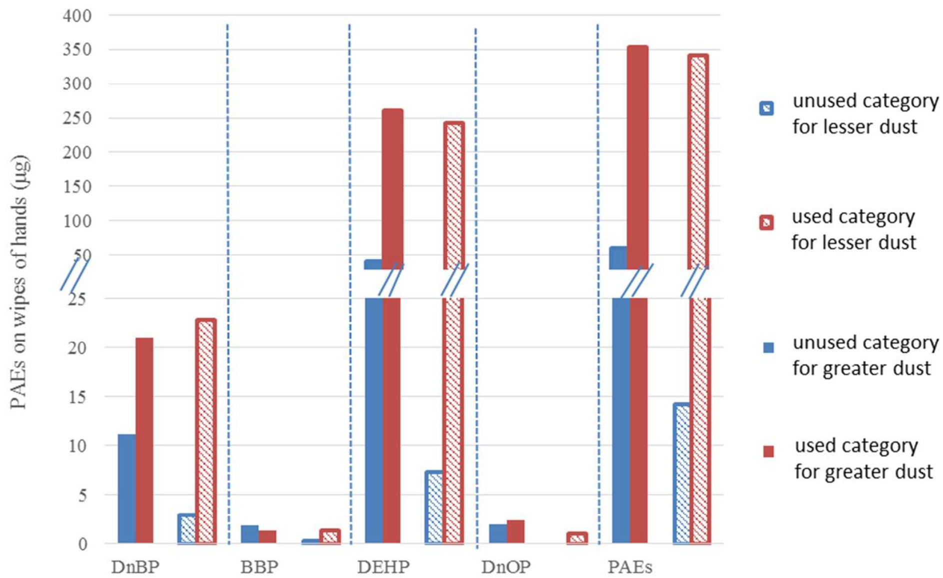

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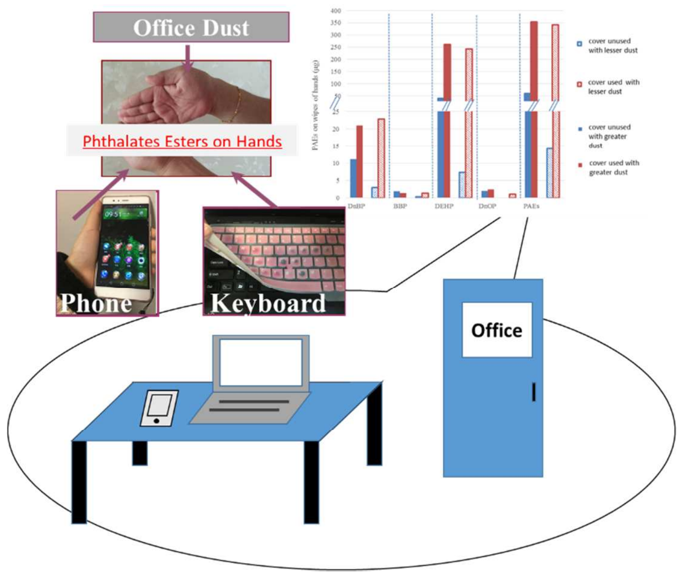
Table

Table 1. Hazard quotients (HQs) for lipophilic phthalate esters (PAEs) through hand to body exposures among categories.

	Individual TDI_i (μg/kg bm/day)	Total absorption through Hand		More frequent hand-washers		Less frequent hand-washers		Not use keyboard dust cover		Use keyboard dust cover	
		50^{th} centile HQ	95^{th} centile HQ	50^{th} centile HQ	95^{th} centile HQ	50^{th} centile HQ	95^{th} centile HQ	50^{th} centile HQ	95^{th} centile HQ	50^{th} centile HQ	95^{th} centile HQ
DnBP	10	1.71×10^{-2}	5.84×10^{-2}	2.31×10^{-2}	2.31×10^{-2}	2.38×10^{-2}	7.59×10^{-2}	1.30×10^{-2}	3.00×10^{-2}	3.72×10^{-2}	9.01×10^{-2}
BBP	200	7.91×10^{-5}	6.46×10^{-4}	8.55×10^{-5}	8.55×10^{-5}	1.17×10^{-4}	1.25×10^{-3}	1.16×10^{-4}	2.72×10^{-4}	1.13×10^{-4}	1.14×10^{-3}
DEHP	50	9.09×10^{-3}	7.82×10^{-1}	1.10×10^{-2}	1.10×10^{-2}	2.99×10^{-2}	1.05	6.98×10^{-3}	2.54×10^{-2}	1.44×10^{-1}	1.06
DnOP	370	2.72×10^{-5}	5.80×10^{-4}	7.12×10^{-5}	7.12×10^{-5}	6.13×10^{-5}	5.64×10^{-4}	1.13×10^{-5}	1.67×10^{-4}	7.09×10^{-5}	6.57×10^{-4}
Sum		2.63×10^{-2}	8.42×10^{-1}	3.42×10^{-2}	3.42×10^{-2}	5.38×10^{-2}	1.13	2.01×10^{-2}	5.58×10^{-2}	1.82×10^{-1}	1.15

TOC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