Pollution Characteristics And Health Risk Assessment of Phthalate Esters In Household Dust In Chengdu, China

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Research Article

Keywords: Phthalate esters, Chengdu, household dust, living habits, health risk assessment

Posted Date: January 27th, 2022

DOI: https://doi.org/10.21203/rs.3.rs-1186935/v1

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Abstract

The characteristics and health risks of 15 phthalate esters in household dust and the association with household attributes were investigated in Chengdu, China. The concentrations of total phthalate esters ranged from 87.9 to 3623 μg/g. Di-(2-ethylhexyl) phthalate (DEHP) was the dominant compound of phthalate esters with a median of 151 μg/g. The statistical analysis result showed household products and synthetic polymer emission were the main sources of phthalate esters. Smoking, cooking, keeping pets and the use of wooden floor and plastic wallpaper might increase the concentrations of phthalate esters in household dust, but opening windows frequently, and increasing the sweeping frequency might cause the decrease of phthalate esters in the indoor environment. The hazard indexes (HI) values of phthalate esters were all below 1, showed no non-carcinogenic for inhabitants. However, the carcinogenic risk of DEHP was $2.54 \times 10^{-6}$, implying the carcinogenic risks via exposure to DEHP from Chengdu household dust were noticeable.

1. Introduction

Phthalate esters are extensively used in various plastic products and consumer products (Gómez-Hens & Aguilar-Caballos 2003, Zhu et al. 2019). In China, exceed 2.1 million tons of plasticizers are produced and phthalate esters contributed to >70% of the annual production in 2014 (Zhu et al. 2019). With the widespread production and applications, phthalate esters are almost everywhere in the environment (Chen et al. 2012). Because phthalate esters do not bind to polymers with chemical bonds, they are easily released into the environment during manufacturing, storage, and usage (Guo & Kannan 2013, Koniecki et al. 2011, Net et al. 2015). The occurrence of phthalate esters in various environmental matrices has been investigated such as urban soil and street dust (0.0002-4.82 μg/g) (Skrbic et al. 2016), surface water (nd-16.10 μg/L) (Luo et al. 2021), sediments (1.69-36.6 μg/g) (Cheng et al. 2019), as well as tobacco cultivation soils (0.22-1.17 μg/g) (Song et al. 2020), food packages (Di-2-ethylhexyl phthalate (DEHP) 0.3-103.33 μg/dm) (Jarošová & Bogdanovičová 2015), and cosmetics and personal care products (nd-25542 μg/g) (Koniecki et al. 2011). In addition, many studies have observed DEHP, di-n-butyl phthalate (DBP) and butyl benzyl phthalate (BBP) and their metabolites may abnormal internal secretions and procreation when long-term exposure, even low concentrations (Hauser & Calafat 2005, Lovekamp-Swan & Davis 2002).

In general, humans spend nearly 90% of their time in indoor environments including home, office, and classroom (Basaran et al. 2020, Xia et al. 2018, Zhu et al. 2019). However, people spend the whole time at home during the special time that staying at home as an effective measure against Novel Coronavirus disease 2019 (COVID-19) by many countries around the world (Chen et al. 2020, WHO 2020). Although we can eventually overcome this virus, it is also necessary to pay more attention to the quality of our home environment. Household dust is a good indicator representing the pollution level of homes (Basaran et al. 2020, Besis & Samara 2012). The occurrence of phthalate esters in household dust from the United States, Italy and Sweden and the megacities in China (e.g., Beijing, Guangzhou, and Shanghai) have been reported (Gevao et al. 2013, Guo & Kannan 2011, Kang et al. 2012, Orecchio et al. 2013). The previous
studies observed there were strong associations between phthalate esters concentration in settled dust and household consumer products such as DEHP-leather polish, di-iso-butyl phthalate (DIBP), BBP, DINP-flooring materials, and BBP-modern window frame (Zhang et al. 2020). Besides, resident personal habits (e.g., smoking, sweeping, cooking), indoor environment and activities (e.g., decoration, the number of residents and pets), characteristics of the building (e.g., age, storey, and height) are also factors that affect the phthalate esters in indoor dust (Wang et al. 2013).

Chengdu is one of the new first-tier cities and the most livable cities in China and a key hub under the Belt and Road initiative (Li et al. 2017, Xu et al. 2018). According to the “2018 World Urbanization Prospects”, there will be 10.7 million population in Chengdu in 2030 (UNDESA 2018). The total area of residential land reached 5282 ha in Chengdu in 2017, including commercial housing (53.28%), housing of limited property rights (23.17%), resettlement housing (21.78%), and affordable housing (1.77%) (Yang et al. 2017). Since 2005, the resettlement housing and affordable housing was increasing, mainly in eastern part of Chengdu (Yang et al. 2017). Affordable housing is the low-cost / leasing of commercial housing mainly provided for low-income urban populations and migrant workers (Man 2011, Shi et al. 2016). Previous studies have shown that low-income population smokes and use chemicals relatively frequently, which may cause poor indoor environmental quality and human health threat (Kolokotsa & Santamouris 2015, Lam et al. 2017). A few studies reported the human health risks of phthalate esters in indoor dust in the USA (Guo & Kannan 2011), Saudi Arabia (KSA) and Kuwait (Albar et al. 2017), and China (Li et al. 2021, Zhu et al. 2019). However, to our best knowledge, no studies are reporting the human health risks of phthalate esters in Chengdu household dust and the relationship between phthalate esters and Chengdu household attributes. The objectives of the present study were: (1) to investigate phthalate esters concentrations and congener profiles in household dust collected from Chengdu; (2) to assess the relationships among phthalate esters in the household dust and building characteristics and resident habits; and (3) to estimate the potential health risks of phthalate esters.

2. Materials And Methods

2.1 Sample collection

Household dust samples were collected by a pre-cleaned small vacuum cleaner from the floor in the living room and bedrooms in 23 households in the downtown of Chengdu (Fig. S1). All samples were sieved to remove large debris, and homogenized with the 154 µm sieve and stored at desiccator until further analysis. A questionnaire was designed to gather more information from the inhabitant to evaluate their living habits, the details of the questionnaire are found in Supporting Information.

2.2 Chemical analysis

Freeze-dried household dust samples (~3g) were spiked with 250 ng of internal standards (DMP-d4, DBP-d4 and DEHP-d4) and equilibrated at room temperature for 3 h. Then, dust samples were extracted with 110 mL acetone/dichloromethane/n-hexane (1:1:1, v/v/v) in a Soxhlet extractor (USEPA 1996a), and the extracts were purified according to florisil cleanup method (USEPA 1996b). Fifteen phthalate esters were
measured from the sample extracts by a GC 6890/MS 5973 with 30 m HP-5MS column (0.25mm diameter and 0.25 mm film thickness). Details about the phthalate esters analyses, QA/QC and calculation are described in the Supplementary material.

2.3 Data analysis

Statistical tests were conducted with SPSS 23.0 for Windows. Kolmogorov–Smirnov test was utilized to examine the normal probability function of the concentration data. Principal component analysis (PCA) and Spearman's correlation coefficient analysis were carried out to identify the possible sources and the relationships between dust-phthalate esters and household factors.

3. Results And Discussion

3.1 Phthalate esters in household dust

The concentrations of phthalate esters in household dust collected from Chengdu are shown in Table S1. Fifteen phthalate esters were detected in household dust samples with a detection frequency from 52.2 to 100%. The concentrations of total phthalate esters varied from 87.9 to 3623 µg/g with a median of 290 µg/g and a mean of 733 µg/g (Table S1). Comparison with other first-tier cities in Eastern China (e.g., Beijing, median 255 µg/g and Shanghai, median 401 µg/g and Guangzhou, median 173 µg/g) (Guo & Kannan 2011) (Fig. 1), there may not be a significant difference of phthalate esters levels in household dust samples between Chengdu and these cities. While lower phthalate esters concentration were observed in Chengdu compared to that found in other first-tier cities in Western China (e.g., Chongqing, median 1751 µg/g and Xi'an, median 1149 µg/g) (Bu et al. 2016, Wang et al. 2014), and that found in other city around the world, such as the USA (median 396 µg/g) (Guo & Kannan 2011), Kuwait (median 2384 µg/g) (Gevao et al. 2013), Italy (mean 1289 µg/g) and Sweden (mean 1055 µg/g) (Orecchio et al. 2013). However, it is worth noting that the extremely high phthalate esters concentrations (exceed 3000 µg/g) also were found in some sampling sites in Chengdu. Thus, for a better understanding of phthalate esters occurrence in Chengdu household dust, more detailed composition profiles and potential sources of phthalate esters are required.

3.2 Composition profiles of phthalate esters

Consistent with that was reported in other studies (Abdi et al. 2021, Guo & Kannan 2011, Zhang et al. 2013), the dominant compound of phthalate esters in all dust samples collected from Chengdu was DEHP, which ranged from 48.2 to 2621 µg/g, with a median of 151 µg/g and a mean of 452 µg/g (Table S1). The concentrations of DEHP were 2-14 times higher than the concentrations of DCHP (median 89.2 µg/g), DBP (median 31.2 µg/g) and DIBP (median 11.5 µg/g), and 100-1000 times higher than other phthalate esters (Fig. 2 and Table S1). DEHP is widely used in plastic products and decoration materials (e.g., floor and wall covering, toys, and adhesives) and can be released into the indoor environment through evaporation, aging, and abrasion (Bornehag et al. 2005, Zhang et al. 2020). While DBP and DIBP also showed relatively higher levels in the household dust, which might be associated with that DBP and
DIBP as the main additives used in personal care products (e.g., cosmetics and pharmaceutical coatings) (Li et al. 2021, Net et al. 2015). In addition, the low levels of other phthalate esters such as DEP and DMP were observed in this study, which is consistent with the reported results from indoor dust samples in other cities (Guo & Kannan 2011, Orecchio et al. 2013), that could be due to the high volatility and high vapor pressure causing the low molecular weight phthalate esters mainly in gas phase than that in dust phase (Basaran et al. 2020, Bergh et al. 2011). Noteworthy, although it’s consistent with the profile in many cities all over the world that DEHP showed the largest proportion in Chengdu household dust (Fig. 1), DEHP concentrations in this study were 10 to 15 times lower than the results reported in previous studies, such as Chongqing, China (median 1543 µg/g) (Bu et al. 2016), and Kuwait (median 2256 µg/g) (Bergh et al. 2011) (Fig. 1). The relatively lower concentrations of DEHP in this study might reflect the trend of decreasing DEHP due to worldwide regulations or restrictions on the use of DEHP (Salthammer et al. 2018, Zhang et al. 2020). In particular, DCHP contributed 30.7% to the total phthalate esters in this study, the result was different from previous reports from China (Beijing, Guangzhou, Shanghai), the USA, and Kuwait (0 to 1.8%) of the total phthalate esters in household dust (Gevao et al. 2013, Guo & Kannan 2011). DCHP is a common plasticizer ingredient for ethyl cellulose, nitrocellulose, vinyl acetate, polyvinyl chloride, and resins, usually be added to the polymers to make these polymers more flexible (Lv et al. 2019, Schecter et al. 2013). As well as DCHP has been detected in many foods and packaging materials (Fierens et al. 2012, Schecter et al. 2013). The reasons underlying the high DCHP Chengdu household dust are unknown at present, but it should be drawn a lot of attention due to growing evidence demonstrated that DCHP had similar potency to DEHP or DBP in the induction of adverse effects on mammalian development and reproductive system (Aydogan Ahbab & Barlas 2013, Li et al. 2016, Lv et al. 2019).

### 3.3 Possible sources of phthalate esters

Principal component analysis (PCA) has been widely used to elucidate the relationship among contaminants and identify their probable sources in various environmental mediums (Cheng et al. 2019, Gu et al. 2018). Kaiser-Meyer-Olkin (KMO) and Bartlett’s sphericity test results were 0.75 and 297.91, respectively (df = 105, p < 0.01), indicating PCA may be a helpful tool in the identification of distinct phthalate esters sources. As shown in Fig. 3, the first-two principal components (PC1 and PC2) were extracted and collectively explained more than 81.0% of the total variance of the phthalate esters in all household dust samples. PC1 (57.4%) was heavily loaded by DBP, DEP, DINP, DAP, DMP, DIBP, DPP and BBP. The lower molecular weight phthalate esters, such as DMP, DEP, DBP and BBP are typically used in cosmetics and personal care products, BBP also widely used in construction materials and home furnishings such as vinyl flooring, adhesives, and synthetic leather (Gómez-Hens & Aguilar-Caballos 2003, Ma et al. 2020, Net et al. 2015). PC2 contributed to 23.6% of the total variance with strong positive loadings of DNOP, DCHP, DEHP, DNP and DEEP. These high-molecular weights phthalate esters were widely used as plasticizers to impart flexibility and general handling properties to PVC and synthetic polymer (Net et al. 2015, Schecter et al. 2013, Zhang et al. 2020). The multiple linear regression (MLR) analysis is performed on the factor scores of PCA, and then quantitative evaluation of the relative
contribution of each identified source of pollutants (Cheng et al. 2019, Ke et al. 2017). In the present study, the regression equations are expressed as follow:

Standardized phthalate esters=0.868 PC1+0.373 PC2 (R²=0.870, p<0.001)

The relative contributions of PC1 and PC2 to the total phthalate esters burden in household dust from Chengdu were calculated from the coefficients, showing PC 1 (personal care products and household chemicals emission) to contribute 69.9%, whereas PC 2 (PVC and other synthetic polymer emissions) contributed 30.0% of phthalate esters in household dust from Chengdu.

In addition, a recent study demonstrated that strong associations between phthalate esters in Chinese household dust and household consumer products such as flooring materials and modern window frame (Zhang et al. 2020). Thus, based on the data from the questionnaire, a total of 13 household factors were defined and calculated the possible relationship with phthalate esters concentrations in Chengdu household dust (Table S2 and Fig. 4). According to the statistical results can identify three categories (e.g., living habits, decorations, and others) of the household factors were found strong associations with dust-phthalate esters (Fig. 4). The living habits included smoking, sweeping frequency, window opening habits, and cooking frequency in this study. Strong positive correlations were observed between smoking and DMP, DEP, and DIBP (p < 0.001), and DBP, DEHP, DINP, and total phthalate esters (p < 0.01), DEEP, DCHP, and DNOP (p < 0.05) (Fig. 4). These results indicated smoking may be one of the most important sources of phthalate esters in household dust in Chengdu. Wang et al. (2020) observed pregnant women exposure the environmental tobacco smoke (ETS) may increase the concentrations of phthalate esters (DIBP, DBP, DPP, and DINP) in their urinary. Song et al. (2020) found the long-term application of plastic film and agricultural inputs (irrigation water and fertilizers) might increase phthalate esters concentrations in tobacco cultivation soils, and then cause the accumulation of phthalate esters in tobacco leaves. In addition, phthalate esters may be released into the household environment from cigarette packaging and printing ink (Gong et al. 2018). Meanwhile, sweeping frequency showed a strong correlation with DMP, DEP, and total phthalate esters (p < 0.01), with DIBP, DBP, DCHP, DINP, and DNP (p < 0.05) (Fig. 4), indicating the higher sweeping frequency to be effective in reducing the accumulation of phthalate esters in household dust. Furthermore, window opening habits also have a strong influence on phthalates esters concentration in the household dust. Pei et al. (2018) found the emission rate of phthalate esters could be increased from the source materials with indoor temperature increasing and the ventilation could reduce the phthalate esters concentration in the dust-phase, so increasing the time to open windows for ventilation and the frequency of house cleaning, which is beneficial to maintaining a good indoor environment. Noteworthy, there were positive correlations among cooking frequency and DEHP, DCHP (p < 0.01), DNOP, and total phthalate esters (p < 0.05), indicated that cooking also was a contributor to phthalate esters in household dust, which could be explained by synthetic condiments and food packaging would be used during the cooking (Fierens et al. 2012, Jarošová &Bogdanovičová 2015, Schecter et al. 2013).
Consistent with previous studies (Ait Bamai et al. 2014, Li et al. 2021, Zhang et al. 2020), strong associations were found between home decorations (e.g., floor cover and wall cover) and phthalate esters in the household dust ($p < 0.05$). The range of associations were observed between floor cover and DCHP, and DEHP ($p < 0.01$), and DEEP, DNOP, and total phthalate esters ($p < 0.05$) (Fig. 4), implying the wooden floors might be found higher concentrations of phthalate esters in the household dust, corroborating wooden floors especially laminated wood floors suspected as the source of phthalate esters (Zhang et al. 2013, Zhang et al. 2020). Plenty of phthalate esters were added as adhesives in the manufacturing process of laminated woods to improve the adhesion property (Ait Bamai et al. 2014, Zhang et al. 2020). In addition, wall covers were associated with DIBP, DEHP, DNOP, DINP, and total phthalate esters ($p < 0.001$), with DMP, DBP, DCHP, and DNP ($p < 0.01$) (Fig. 4), the results might provide further evidence that plastic wallpaper and latex paints were the important sources for phthalate esters in the household dust (Shinohara et al. 2019).

The results also showed that positive correlations between pets and phthalate esters in this study (Fig. 4). The high-molecular weights phthalates esters (e.g., DEHP and DINP) are restricted in children's toys and child-care articles in the US, Europe and China, but there are no restrictions on pet toys (Wooten & Smith 2013). Many previous studies had reported the detection of high-molecular weights phthalate esters in pet toys, such as, concentrations of DEHP (4 out of 13) and DINP (10 out of 13) in 13 pet toys ranged from 6.9 to 54% (v/v) in Denmark, and 5 out of 6 phthalates were detected in canine toys and training devices in the US (Müller et al. 2006, Wooten & Smith 2013). Notably, the correlations between DCHP and household factors were similar to that of DEHP, indicating DCHP not only had similar potency to DEHP in the induction of adverse effects on the reproductive system, but also might have similar sources in the indoor environment (Aydogan Ahbab & Barlas 2013, Lv et al. 2019).

In this study, although non-significant correlations were found between building characteristics (e.g., floor level, age of building, and area of building) and phthalate esters in the household dust, previous studies found that phthalate esters might accumulate especially high-molecular weights phthalate esters in household dust in the older house since the erosion and abrasion of building materials with time (Bornehag et al. 2005, Shinohara et al. 2019). In addition, the high-rise and large dwelling-size houses have a stronger inhibitory effect on the accumulation of phthalate esters in household dust. The high-rise building causing the pollutants (e.g., heavy metals, polycyclic aromatic hydrocarbons, and phthalate esters) loaded in the particulates would be difficultly resuspended in vertical directions, and large size house enhancing horizontal diffusion space of pollutants, resulting in the decrease of pollutants concentrations in the indoor environment (Bornehag et al. 2005, Hang et al. 2012).

### 3.4 Human health risks of phthalate esters

To estimate the risks of phthalate esters exposure in household dust from Chengdu, the average daily dose (ADD, $\mu$g/kg-bw/day) of DMP, DEP, DIBP, DBP, BBP, DEHP and DNOP via ingestion, inhalation, and dermal absorption pathway were estimated for children and adults (Table 1). The total ADD of phthalate esters from household dust for children was about 7.5 times higher than adults, implying children were more susceptible to phthalate esters intake than adults. Similar results were observed in previous studies.
(Albar et al. 2017, Li et al. 2021, Zhu et al. 2019). The ADDs of phthalate esters for children and adults via dust ingestion were 2-7 orders of magnitude higher than dermal adsorption and dust inhalation, indicating the household dust ingestion was the principal pathway for inhabitant exposure to phthalate esters. The results were consistent with previous reports (Li et al. 2021, Zhu et al. 2019).
Table 1
Risk assessment of human exposure to phthalate esters via household dust.

<table>
<thead>
<tr>
<th></th>
<th>DMP</th>
<th>DEP</th>
<th>DIBP</th>
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<td>1.03E-03</td>
<td>5.69E-02</td>
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<td>9.39E-03</td>
<td>1.83E+00</td>
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Note, ADDing, ADDinh, and ADDder indicates the average daily dose (µg/kg-bw/day) of individual PAEs via ingestion, inhalation, and dermal absorption exposure; HQing, HQinh, and HQder suggests hazard quotient of individual PAEs by ingestion, inhalation, and dermal absorption exposure; HI represents hazard index; LADD denotes the lifetime average daily exposure doses of the carcinogenic PAEs; CR is carcinogenic risk. Bold indicates the values above $1 \times 10^{-6}$, which might pose health risk to inhabitants.
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</table>

Note, ADDing, ADDinh, and ADDder indicates the average daily dose (µg/kg-bw/day) of individual PAEs via ingestion, inhalation, and dermal absorption exposure; HQing, HQinh, and HQder suggests hazard quotient of individual PAEs by ingestion, inhalation, and dermal absorption exposure; HI represents hazard index; LADD denotes the lifetime average daily exposure doses of the carcinogenic PAEs; CR is carcinogenic risk. Bold indicates the values above $1 \times 10^{-6}$, which might pose health risk to inhabitants.

Furthermore, the values of HI for all phthalate esters were all below 1 suggesting that no non-carcinogenic risks of phthalate esters in household dust were found for Chengdu inhabitants (Table 1). The HI values of individual phthalate ester in the household dust samples in the decreasing order as follows: DEHP > DBP > DIBP > BBP > DNOP > DEP > DMP (Table 1). The carcinogenic risk for human exposure to BBP and DEHP in the household dust was $1.77 \times 10^{-9}$ and $2.54 \times 10^{-6}$, respectively (Table 1), which cancer risk of DEHP was in the range of $10^{-6}$ to $10^{-4}$, indicating that there might be a potential carcinogenic risk of DEHP in household dust in Chengdu.

### 4. Conclusions

The present study investigated the concentrations of phthalate esters in household dust from Chengdu. DEHP was the largest proportion of phthalate esters in this study, but its concentrations were 10 to 15 times lower than the results reported in previous studies. Besides, DCHP contributed 30.7% to the total phthalate esters in the present study, the result was different from previous reports from China. Living habits (smoking, sweeping frequency, window opening habits, and cooking frequency), home decorations (floor cover and wall cover) and other factors such as pets can significantly affect the concentrations of phthalate esters in household dust in Chengdu. Particularly, the inhabitants in Chengdu might face the potential carcinogenic risk of phthalate esters in household dust. Thus, some suggestion to the general population in Chengdu, (1) Smoking less and opening windows frequently to maintain the indoor air circulation; (2) The pet supplies need to be stocked in a fixed place; (3) Increasing the sweeping frequency of indoor environment, especially the older houses.

### Declarations


**Funding** The research works of the above study were funded by the National Natural Science Foundation of China (No. 21507095), and Sichuan Provincial Youth Science and Technology Fund (No.2017JQ0035).

**Data availability** All data generated or analyzed during this study are included in this published article.
Ethics approval and consent to participate The manuscript did not contain any reporting studies involving human data.

Consent for publication Themanuscript does not contain any individual person data.

Conflict of interest The authors declare no competing interests.

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Figures

Figure 1

Concentrations (μg/g) of phthalate esters in household dusts in Chengdu and other cities around the world.
Figure 2

The concentration (μg/g) of individual phthalate esters in household dusts from Chengdu.
Figure 3

Principal component analysis of phthalate esters in the household dusts.
Figure 4

Relationships between the household factors and phthalate esters concentrations in the household dust.

Supplementary Files

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