Associations of phthalate concentrations in floor dust and multi-surface dust with the interior materials in Japanese dwellings

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Abstract

Phthalates are widely used as plasticizers in numerous products. However, there has been some concern about the various effects they may have on human health. Thus, household phthalate levels are an important public health issue. While many studies have assessed phthalate levels in house dust, the association of these levels with building characteristics has scarcely been examined. The present study investigated phthalate levels in house dust samples collected from the living areas of homes, and examined associations between these phthalate levels and the interior materials. Dust was collected from two portions of the living area: floor dust from the entire floor surface, and multi-surface dust from objects more than 35 cm above the floor. The levels of seven phthalates were measured using gas chromatography/mass spectrometry in selective ion monitoring mode. Phthalate levels were higher in multi-surface dust than in floor dust. Among floor dust samples, those from dwellings with compressed wooden flooring had significantly higher levels of di-iso-butyl phthalate compared to those with other floor materials, while polyvinyl chloride (PVC) flooring was associated with higher di-2-ethylhexyl phthalate (DEHP) levels. Among multi-surface dust samples, higher levels of DEHP and di-iso-nonyl phthalate (DINP) were found in samples from homes with PVC wallpaper than without. The number of PVC interior materials was significantly positively correlated with the levels of DEHP and DINP in multi-surface dust. The phthalate levels in multi-surface dust were associated with the interior surface materials, and those in floor dust were directly related to the flooring materials. Our findings show that when using house dust as an exposure assessment, it is very important to note where the samples were collected from. The present report provides useful information about the association between phthalates and dust inside dwellings, which will assist with establishing public health provisions.

Keywords:

Phthalates

DEHP

PVC

House dust

Interior materials

Abbreviations:

BBzP, benzyl butyl phthalate

BHT, dibutylhydroxytoluen

DBP, dibutyl phthalate

DEHA, di-(2-ethylhexyl) adipate

DEHP, di-2-ethylhexyl phthalate

DEP, diethyl phthalate

DiBP, di-iso-butyl phthalate

DINP, di-iso-nonyl phthalate

DMP, dimethyl phthalate

DnBP, di-n-butyl phthalate

LOD, limit of detection

LSM, least square mean

PVC, polyvinyl chloride

SVOC, semi-volatile organic compounds

1. Introduction

Phthalates are semi-volatile organic compounds (SVOC) that have been used as plasticizers for various plastic products, such as toys, food containers, furniture, personal care products, wallpaper, flooring materials, cable, artificial leather, glue, and paint. Phthalates are slowly released from these products, and are partitioned among the gas phase, airborne particles, and settled dust. The most commonly used phthalate is di-2-ethylhexyl phthalate (DEHP), which accounts for about 50% of plasticizers and about 65% of phthalates in Japan (Japan Plasticizer Industry Association and Ministry of Economy, 2003).

In the late 1990s, the endocrine disrupting effects of phthalates became a matter of international concern (Gray et al., 1982; Oishi et al., 1993). In Europe, the USA, and Japan, this led to regulation of the use of several phthalates in PVC-containing toys intended to be placed in the mouth by children (EU. Directive, 2005; U.S. Consumer Product Safety Improvement Act, 2008; Japan Ministry of Health and Welfare, 2002), as well as in food containers that may touch oily food (EU Commission Directive, 2007; Japan Ministry of Health and Welfare, 2002; U.S. FDA CFR178.3740). While the main source of phthalate exposure has traditionally been thought to be ingestion, phthalates have also been detected in residential indoor air and house dust (Adibi et al., 2003; Rudel et al., 2003; Wormuth et al., 2006). Since phthalates are not chemically bound to products, they can diffuse within the materials, leak out, and then disperse in air or adhere to dust (Fujii et al., 2003). Therefore, phthalates might easily penetrate into house dust that settles on phthalate-containing products (Seto and Saito, 2002). Since our lives are surrounded by products that contain phthalates, the potential for considerable health problems exists. However, the laws regulating phthalate use only apply to food containers, medical devices, and vinyl toys that could be placed in a child’s mouth. Regulation of phthalate use in building materials and interior materials should also be considered.

Several studies have reported the phthalate levels inside buildings. The interior materials, such as flooring, wallpaper, and ceiling materials, are thought to affect the indoor phthalate levels (Bornehag et al., 2005; Clausen et al., 2003; Fromme et al., 2004; Jaakkola and Knight, 2008; Jaakkola et al., 1999; Weschler and Nazaroff, 2010; Xu et al., 2009, 2010). In particular, PVC flooring, PVC wallpaper, and polishing agents have been associated with high DEHP levels in house dust (Bornehag et al., 2005;

Kolarik et al., 2008a). Lower proportions of plastic materials and carpeting are associated with lower total phthalate level (Abb et al., 2009). High levels of DEHP in PVC and in house dust are known to be an important source of phthalate exposure. Regarding the adverse effects of phthalates, plastic interior surfaces, signs of dampness-related DEHP degradation, and higher levels of DEHP in house dust are related to bronchi problems, wheezing, and asthma in children (Bornehag et al., 2004; Jaakkola et al., 1999; Kolarik et al., 2008b; Larsson et al., 2010; Norback et al., 2000).

Measuring phthalate concentrations in house dust is a widely used method for estimating indoor phthalate levels. However, only four studies have investigated the associations between residential characteristics and phthalate levels in house dust. Of these studies, two studies collected dust from objects more than 35 cm above the living room floor (Bornehag et al., 2005; Kolarik et al., 2008a), and two collected dust from the floor (Abb et al., 2009; Kang et al., 2012). No study has collected dust from both the floor and above the floor. Furthermore, the contributions of various sources of phthalates in house dust and the association between the levels of phthalates in house dust and the interior materials remain unknown.

The present study aimed to evaluate the phthalate levels in house dust from different sampling places, and to examine the associations between interior materials (such as flooring, wallpaper, and ceiling materials) and the phthalate concentrations in the house dust.

2. Material and methods

2.1. Study population

This study was conducted in two phases: a baseline questionnaire survey in 2008 and a questionnaire, environmental measurements, and building investigation survey conducted between 2009 and 2010. The results of the baseline questionnaire survey have been previously reported (Ukawa et al., 2012). Briefly, all 6393 school children from 12 public elementary schools in Sapporo were asked to participate in the study, of which 4408 children responded to the questionnaire (response rate 69.0%). The baseline questionnaire included questions about personal and dwelling information. Personal information included questions on gender, school grade, allergies, number of siblings, number of family members, and parental history of allergies. To define children's allergies, the International Study of

Asthma and Allergies in Childhood (ISAAC) core questionnaire (The ISAAC Steering Committee 1998) was used. Dwelling information included questions about type of dwelling, building structure, building age, renovation, wall-to-wall carpeting, heating system, indoor smoker at home, pet keeping, and dampness-related signs, such as mold growth, moldy odor, condensation, and water leakage. A total of 951 children (832 families) agreed to allow a home visit to conduct environmental measurements. In 2009 and 2010, we contacted children who were still attending the same elementary school as in 2008, excluding those who left blanks on the baseline questionnaires regarding their gender, grade, or SBS (Sick Building Syndrome) and allergies for ISAAC (International Studies of Asthma and Allergies on Childhood). This selection procedure identified a total of 128 families who allowed home visits for environmental measurements, dust collection, and questionnaire survey in October and November of 2009 and 2010.

2.2. Questionnaire

Self-administered questionnaires were distributed and collected by the investigators when they visited each house for dust sampling in 2009 and 2010. The questionnaire included questions about the type of dwelling, building structure, age of building, residence years, renovation, annual household income, and dampness-related signs, such as mold growth, moldy odor, condensation, water leakage, and high bathroom humidity.

2.3. Environmental measurements

In 128 dwellings, indoor environmental measurements were performed by well-trained investigators in a main living area where all inhabitants commonly spent most of their time. The Thermo Recorder TR-72U (T & D Corporation, Nagano, Japan) was used to monitor room temperature and relative humidity in each house for 48 h.

2.3.1. Phthalate concentrations in settled dust

Dust samples were collected using a previously reported strategy (Kanazawa et al., 2010). Briefly, dust samples were categorized as one of two types: floor dust or multi-surface dust. Floor dust samples were collected from the floor surface and from objects within 35 cm above the floor. Samples of multi-surface dust were collected from the surfaces of objects that were more than 35 cm above the floor including shelves, cupboards, moldings, frames, door frames, windowsills, TV sets, audio sets, personal computers, and interior materials such as wallpaper and the ceiling. The same type of hand-held vacuum cleaner (National HC-V15, Matsushita Electric works, ltd., Osaka, Japan; 145W) equipped with a paper dust bag (Nichinichi Pharmaceutical Co., Ltd., Mie, Japan) was used at all dwellings. The collected dust was weighed after the removal of unwanted substances, such as human and animal hair, insects, food scraps, scrap paper, etc. Samples were stored in stoppered glass test tubes that were sealed with fluoric-tape, wrapped with aluminum foil, and kept at −20 °C in until the day of analysis.

The collected dust was subjected to ultrasonic extraction with residue analysis-grade acetone (Wako Pure Chemical Industries, Ltd., Osaka, Japan) for 15 minutes, and sterilized at 250 °C for 2 hours. Gas chromatography/mass spectrometry (GC/MS) in SIM mode was used to analyze the concentrations of seven phthalates—dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (DnBP), di-iso-butyl phthalate (DiBP), benzyl butyl phthalate (BBzP), DEHP, and di-iso-nonyl phthalate (DINP)—as well as di-(2-ethylhexyl) adipate (DEHA) and dibutylhydroxytoluen (BHT). These analyses were conducted at the Tokyo Metropolitan Institute of Public Health and Osaka Occupational Health Service Centre, Japan Industrial Safety and Health Association. The analysis methods have been previously described (Kanazawa et al., 2010). Background phthalate levels were measured from the vacuum cleaner and the filter, verifying that high phthalate concentrations were not detected from these items.

2.4. Investigations of dwelling characteristics

In the main living area, we investigated the floor area (m^2), ceiling height (cm), frequency of cleaning the living room floor (times/week), and interior materials of the floor, walls, and ceiling. There were four categories of floor materials: PVC floor, compressed wooden floor, wall-to-wall carpet, and tatami/tiles/natural wooden floor. Wall and ceiling materials were categorized as PVC or not PVC. Non-PVC walls included paint, concrete, and wood, while non-PVC ceilings included wood and plaster board. Based on the number of areas where PVC interior materials were used, dwellings were given a numeric score of 0–3, with 0 indicating no PVC in the floor, walls, and ceiling, and 3 indicating PVC use in all three surfaces. Inhabitants were asked about the most recent date of cleaning the living area, and the dates of dust accumulation were calculated. A pre-established checklist was used to reduce the subjectivity of the investigations.

2.5. Quality control and quality assurance

Recovery tests were performed using dust samples. After 50 ng of each phthalate was individually added to 50 mg dust samples, the air-dried samples were extracted with 1 ml of acetone and analyzed by GC/MS (n =3). Recovery rate ± standard deviations ranged from 80.5 ± 1.6 for DMP to 99.9 ± 4.5 for DINP (data not shown). The instrumental limit of detection (LOD) was defined as the absolute amount of analyte that yielded a signal-to-noise ratio of 3 (S/N =3). As for DnBP and DEHP which were detected in method blanks, LOD was calculated from ten-fold of the standard deviation (10SD) which was calculated from the blank test (n=6). The calculated LOD for each phthalate in the dust is shown in Table 3; if phthalate concentrations were below the LOD, they were assigned a value of half the LOD. A phthalate was identified when its peak was within ±5 seconds of the retention time of a specific phthalate in the calibration standard and the relative noise intensity was within ±20% of that of the standard phthalate. Quantification of each phthalate was first determined based on the peak area ratio of the standard curve, and then the concentrations of individual phthalates in the dust samples (Cd) (μg/g) were calculated based on Equation 1:

$$Cd = [(As - At) \times E] \div (v \times W) \quad (1)$$

where As is the sample weight injected for GC/MS (ng), At is the weight of the travel blank injected for GC/MS (ng), E is the extract volume (ml), v is the injected volume (μl), and W is the weight (g) of the dust sample that was used for extraction. To avoid phthalate contamination, all glass tubes and stainless steel equipment for sample collection and analysis were ultrasonicated for 10 min in acetone, rinsed with acetone, and then air dried. To examine the background levels of phthalates from materials used for sampling, the vacuum dust bag and the ethanol-soaked cotton used to wipe the vacuum nozzle were extracted with acetone and analyzed by GC/MS to confirm that there were no phthalate peaks (data not shown).

2.6. Data analysis

The phthalate concentration in dust and the building characteristics were analyzed for all 128 homes visited. The data for phthalate concentrations were not normally distributed according to the Shapiro-Wilk W-test. The correlation coefficient values between floor and multi-surface dust were calculated using the Spearman's rank correlation test. Phthalates levels in floor and multi-surface dust were compared using the Wilcoxon matched rank test. Correlation coefficient values between phthalate levels and continuous variables such as building age, floor area, height of ceiling, frequency of cleaning living room, temperature, and humidity were calculated using Spearman's rank correlation test. Potential associations between dust phthalate concentrations and building characteristics were analyzed using the nonparametric Mann-Whitney U test. Associations between phthalate concentrations and flooring materials were analyzed by Kruskal-Wallis test. The phthalates that showed significant associations with flooring materials were further analyzed using multiple comparisons, and the *P* values were adjusted using Bonferroni's correction. The trends relating to the number of PVC materials and phthalate levels were analyzed using the Jonckheere-Terpstra test and multiple comparison tests. Multiple comparison tests were conducted to set up "Numbers of PVC materials = 0" as the control. Multivariate regression analyses were conducted in those phthalates that showed significant associations in table 4. Adjustments were made for dampness index and household income. The dampness index (0-5) was calculated by summing dampness related signs, including condensation, visible mold, moldy odor, water leakage, and high air humidity in the bathroom. Levels of phthalates are presented as least square mean (LSM). For statistical analyses, a two-tailed test and a 5% level of significance were used. All analyses were performed using SPSS 19 for Macintosh (SPSS Inc., Chicago, IL, USA).

2.7. Ethical considerations

All participants gave their written informed consent. The study protocol was approved by the ethical board for epidemiological studies at Hokkaido University Graduate School of Medicine and at all the regional universities involved in the study.

3. Results

Table 1 shows a comparison of the dwellings in the present study with those in the 2008 questionnaire survey; the dwellings in the present study were newer, but had higher dampness signs. Table 2 shows the characteristics of building in the present study. Of all investigated dwellings, 71.9% had compressed wooden flooring, 88.3% used PVC wallpaper, and 85.9% used PVC ceiling. The dwelling type and building structure were almost evenly divided between single-family house and multi-family apartment, and wooden structure and reinforced concrete structure, respectively.

Table 3 shows the phthalate distribution in dust. DEHP was found at the highest concentration in both floor and multi-surface dust. DEHP and DINP were each found in 100% of floor dust samples, followed in frequency by DnBP (95.3%), DiBP (93.0%), and BBzP (68.0%). In multi-surface dust, DINP and DiBP were the most-frequently detected (100%), followed by DEHP (99.2%), DnBP (97.7%), and BBzP (85.2%). DMP and DEP were not detected in more than half of the samples of both floor and multi-surface dust, and thus were excluded from further analysis. Compared to floor dust, multi-surface dust had significantly higher concentrations of all phthalates (DnBP: Z = -2.62, P = 0.009; DnBP: Z = -4.25, P < 0.001; BBzP: Z= -4.36, P < 0.001; DEHP: Z = -5.46, P < 0.001; DINP: Z= -4.06, P < 0.001). We found significant positive correlations between the phthalate concentrations in floor dust and multi-surface dust (DiBP: ρ = 0.293, P < 0.001; DnBP: ρ = 0.206, P = 0.02; BBzP: ρ = 0.263, P = 0.003; DINP: ρ = 0.258, P = 0.003), except for DEHP.

Table 4 shows the association between building characteristics and phthalate concentrations in dust. In the case of floor dust, the type of floor material was significantly associated with levels of DiBP (P = 0.004) and DEHP (P = 0.001). The median DiBP level was highest in homes with compressed wooden floor, followed by tatami/tiles/natural wooden floors, whereas that of DEHP was highest with PVC floor. Household income was associated with levels of DEHA (P =0.017). For multi-surface dust, the DEHP and DINP levels were significantly higher in homes with PVC wallpaper than without (DEHP: P = 0.031; DINP: P = 0.004). Single-family houses and wooden houses had significantly higher median DEHP concentrations than multi-family houses (P = 0.014) and reinforced concrete structures (P = 0.014), respectively. Ceiling height was negatively correlated with the levels of DnBP (ρ = -0.201, P = 0.023) and DEHP (ρ = -0.253, P = 0.004). Dampness index was associated with levels of DnBP (P

=0.001), DEHP (P =0.022), and BHT (P =0.014). In both floor and multi-surface dust, DnBP (Floor: ρ = 0.241, P = 0.006; Multi-surface: ρ =0.460, P < 0.001) and DEHP (Floor: ρ = 0.235, P = 0.007; Multi-surface: ρ =0.180, P = 0.042) were significantly positively correlated with building age. Floor area (m^2), temperature (℃), and humidity (%) were not associated with either floor or multi-surface dust (data not shown).

Fig. 1 shows multiple comparisons of the concentrations of DiBP and DEHP in floor dust with different floor materials. Compared to wall-to-wall carpet, compressed wooden floor was significantly associated with a higher median DiBP concentration (P = 0.003). PVC flooring was associated with a significantly higher median DEHP concentration than compressed wooden flooring (P = 0.001), wall-to-wall carpet (P = 0.017), and other types of flooring (P = 0.001). Fig.2 shows multivariate regression analyses of concentrations of DiBP (a) and DEHP (b) in floor dust and different flooring materials. After adjustment for dampness index and household income, compressed wooden floors had significantly higher levels of DiBP than wall-to-wall carpet (P = 0.002) or tatami/tiles/natural wooden floors (P = 0.005). PVC floor also had higher levels of DiBP than wall-to-wall carpet (P = 0.010). However, after adjustment for Bonferroni's correction, only the association between PVC floor and tatami/tiles/natural wooden floors remained statistically significant. PVC floor had higher levels of DEHP than compressed wooden floor (P <0.001), wall-to-wall carpet (P = 0.001), or tatami/tiles/natural wooden floors (P <0.001). After adjusting for Bonferroni's correction, statistical significance remained. Fig. 3 shows that the concentrations of DEHP and DINP in multi-surface dust were each significantly positively associated with the numbers of PVC interior materials (P = 0.012 and P = 0.005, respectively). When comparing "numbers of PVC materials = 0" as the control to the others, significant differences were obtained in the numbers of PVC materials = 2 (P = 0.026) and 3 (P = 0.030) in DEHP and the numbers of PVC materials = 2 (P = 0.028) in DINP. However, after adjustment for dampness index and household income, the only significant relationship that remained was the number of PVC materials = 2 (P =0.007) in DINP (Fig.4).

4. Discussion

In the present study, we found that homes with PVC flooring had higher DEHP levels in floor dust (P = 0.001). Additionally, dwellings with PVC wallpaper had significantly higher levels of DEHP in multi-surface dust compared to in homes with other wall materials (2400 μ g/g dust vs. 888 μ g/g dust, P = 0.031). Dwellings with PVC ceilings tended to show higher DEHP levels in multi-surface dust compared to in homes with other ceiling materials. Bornehag et al. (2005) also reported that DEHP concentration in house dust was associated with the amount of PVC flooring in the house. Since phthalates are not chemically bound to products, DEHP can escape from PVC, and adsorb to the floor or to object surfaces (Fujii et al., 2003) and can then adsorb to dust that collects on these surfaces (Seto and Saito, 2002). The presently identified association between DEHP level in dust and PVC materials in the home supports the findings of previous reports (Bornehag et al., 2005; Fujii et al., 2003; Seto and Saito, 2002).

DINP is used as an alternative to DEHP; therefore, its use as a PVC plasticizer is very similar to the use of DEHP (Kavlock et al., 2002a). Compared to homes with other wall and ceiling materials, dwellings with PVC wallpaper and ceilings showed significantly higher DINP levels in multi-surface dust. We found that the median concentrations of DEHP and DINP in multi-surface dust were significantly higher with each increase in the number of PVC materials in the main living area (DEHP: P = 0.012, DINP: P = 0.005; Fig. 3). Most Japanese dwellings use PVC materials in both the walls and ceiling, meaning that in the average room, a person is surrounded by at least three PVC surfaces (one ceiling and two wall surfaces). Therefore, the high levels of DEHP and DINP in multi-surface dust may be affected by PVC use in wallpaper and ceilings.

We further found that higher DiBP levels in floor dust were associated with compressed wooden floor (butcher-block, parquet) (P = 0.003), which was the most common floor material in this study. Compressed wooden flooring is usually made from 4–5 thin pieces of compressed wood, which are glued together and covered with wax, paint, and sometimes flame retardants on the surface. The applied gloss agents, plastic additives, paint, and varnish contain DiBP (European Commission, 2004), and Kolarik et al. (2008a) reported that compressed wooden flooring contains phthalates. As compressed wooden floor is the most common flooring material in Japanese dwellings, DiBP exposure is a concern. Here we found that DiBP concentration in floor dust was positively correlated with the frequency of cleaning the living room (ρ = 0.197, P = 0.027), which may be because more frequent cleaning is associated with a greater frequency of using DiBP-containing cleaning products. Afshari et al. (2004) reported that using polyolefin covered with wax for floor polishing increased di-butyl phthalate (DBP) concentration in chamber air by two-fold.

Dwellings with PVC ceilings had significantly lower DnBP levels in both floor and multi-surface dust compared to those with other ceiling materials (P =0.010, P = 0.038, respectively). In the dwellings with PVC walls, DnBP levels in floor and multi-surface dust also tended to be lower than in those with other wall materials. While more than 95% of the DEHP and DINP produced is used as a PVC plasticizer (European Commission, 2003; European Commission, 2008; Japan Ministry of Environment, 2002; U.S. Department of Health and Human Services, 2001), DnBP is mainly used as a coalescing aid in latex adhesives, as well as a plasticizer in cellulose plastics and a solvent for dyes (Japan Ministry of Environment, 2002; Kavlock et al., 2002b). Our results suggest that DnBP may be predominantly contained in materials such as paint, paper, and wooden wall or ceilings, rather than in PVC materials. Like DEHP, DiBP and DnBP have been characterized as environmental pollutants. Animal studies have reported that DBP has anti-androgenic activity in male rats (Adibi et al., 2003; Gray et al., 2006; Mylchreest et al., 1998, 1999, 2000; Nagorka et al., 2011), and that DBP has allergy adjuvant effect in mice (Larsen et al., 2002). Thus, the human health impacts of using DiBP in homes must also be considered.

Older buildings were associated with higher concentrations of DnBP (ρ = 0.241, P = 0.006; ρ = 0.460, P < 0.001, respectively), and DEHP (ρ = 0.235, P = 0.008; ρ = 0.180, P = 0.042, respectively), in both floor and multi-surface dust, and of BHT (ρ = 0.227, P = 0.010), in multi-surface dust. Bornehag et al. (2005) also reported that older buildings had higher concentrations of DEHP than buildings from later periods, and that the total DEHP concentration in Sweden has decreased over recent years. Japan developed guidelines for DnBP in indoor air in 2000, and guidelines for DEHP in 2001, which led to reduce phthalate usage. The Japan Plasticizer Industry Association (2011) reported the usage of 2521 tons of DnBP in 2008, and 1531 tons in 2011, while 162,520 tons of DEHP were used in 2008, and 128,772 in 2011. This may imply that compared to newer homes, older dwellings incorporate higher levels of DnBP and DEHP in the interior materials, furniture, and so on. Building age was also significantly negatively associated with ceiling height (r = −0.400, $P < 0.000$; data not shown), and ceiling height was significantly negatively correlated with the levels of DnBP (ρ = -0.201, $P = 0.023$), DEHP (ρ = -0.253, $P = 0.004$), and BHT (ρ = -0.179, $P = 0.042$) in multi-surface dust. However, after accounting for the partial correlations between each phthalate (DnBP, DEHP, and BHT), ceiling height, and building age, these associations were no longer significant, indicating that building age is a confounding factor of the association between the ceiling height and phthalate levels.

Dwelling characteristics were influenced by socio-economic status (SES). In addition, several previous studies have shown that signs of dampness-related DEHP degradation was associated with health problems (Norback et al., 2000; Bonehag et al., 2004). In this study, high dampness index was related to older houses ($P = 0.001$), low household income ($P = 0.004$), and rented houses ($P < 0.001$). Low household income was also related to rented houses ($P < 0.001$) (data not shown). Furthermore, levels of DEHP in multi-surface dust significantly increased according to dampness index ($P = 0.022$). From these results, since dampness-related problems, SES, and older houses were identified as confounding factors for phthalates levels, we conducted additional analyses to further investigate the associations between phthalates levels and dwelling characteristics, including SES and dampness-related problems (Fig. 2 and 4). Significant associations between compressed wooden floor and higher levels of DiBP in floor dust, and that of PVC floor and higher levels of DEHP in floor dust remained after adjustment for dampness index and household income. Levels of DnBP, DEHP, and BHT were related to building age, however, after adjustment for dampness index and household income in multivariate linear regression analyses, only DEHP in floor dust remained statistically significant (157 μg/g dust/ 5 years, $P = 0.011$: data not shown). Even after adding building age as a confounding factor to the adjusted model, levels of DEHP were still significantly higher in PVC floors than each of the other materials ($P < 0.017$; data not shown). In this study, we concluded that having PVC flooring was the greatest contributing factor for high DEHP levels in floor dust, and this association was independent of dampness-related problems, low-SES or older building age. As for high DnBP levels in floor dust, covering the floor with compressed wood was this greatest contributing factor and this association was independent of having dampness-related problems or low-SES. On the other hands, some significant associations disappeared in the adjusted model. In this case, levels of phthalates and dampness and/or SES were strongly associated with each other.

We observed significantly higher phthalate concentrations in multi-surface dust than in floor dust ($Z = -7.09 \sim -2.62$, $P < 0.01$). There are two explanations for this finding. First, multi-surface dust was collected directly from the surface of many kinds of plastic products, such as children's toys, electrical appliances, window panels, and furniture. Even non-plastic products—such as molding, picture frames, and window panels—contain phthalates in glue, paint, and gloss agents (Afshari et al., 2004; Xu et al., 2010). In addition, as for electrical appliances, it is well known that phthalates leak from electrical devises when they are warm. Thus, phthalates leaking from electrical devises to multi-surface dust may also be a reason for higher phthalates concentrations in multi-surface dust. However, in this study, since we collected multi-surface dust from plastic products, non-plastic products, electrical devises, and non-electrical devises all at the same time, we could not consider each association separately. Therefore, further studies are needed to clarify this hypothesis. Secondly, the living room floors tended to be wiped or vacuumed every two days on average, but furniture was cleaned much less frequently (data not shown), meaning that multi-surface dust samples remained for longer periods of time than floor dust samples (Lioy et al., 2002). This may indicate that sampling dust from multiple surfaces of household products could be used to assess phthalate levels over long time periods, while sampling floor dust could be used to assess phthalate levels over short time periods. Furthermore, when using house dust as an exposure assessment, it is very important to note where samples were collected from.

This is the first study to report the relationship between Japanese dwelling characteristics and the phthalate concentrations in both floor and multi-surface dust. Compared with those in our 2008 baseline questionnaire survey (n = 4408), the dwellings selected for home environmental measurements in the present study were newer buildings and showed a higher prevalence of dampness signs; the proportions of different dwelling types and building structures did not differ. It is possible that those who were more interested in home environments chose to participate in the present study.

Phthalates are added to enormous numbers of consumer products for indoor use, but the usage and intended use of phthalates differ among countries. Other studies have previously reported the phthalate concentrations in house dust (Table 5). In the present study, we found higher DEHP concentrations in multi-surface dust than reported elsewhere (Bornehag et al., 2005; Kolarik et al., 2008; Langer et al., 2010; Hsu et al., 2012). PVC wallpaper is the most commonly used wall material in Japanese dwellings (88.3%), while it is used in only 9.5% of homes in Sweden (Bornehag et al., 2005). On the other hand, the BBzP concentrations in both floor and multi-surface dust in this study were lower than in other studies. BBzP is not a principal phthalate product, with less than 1000 tons produced and/or imported per year, and it is mainly used as a plasticizer for wall and floor tiles, paint, and adhesives (Japan Plasticizer Industry Association and Ministry of Economy, Trade and Industry, 2003). On the other hand, in Europe, although the market for BBzP has decreased over the last decade, 19,500 tons of BBzP were produced in 2004, and its main intended use is for PVC flooring (European Commission, 2007). PVC flooring is used in 52.0% of homes in Sweden (Bornehag et al., 2005), and was used in 7.8% of homes in the present study. Thus, the observed differences in BBzP concentration between countries are likely due to the different phthalate usage.

Here we measured the phthalate concentrations in both floor and multi-surface dust. Several other studies have collected dust from above floor level, such as from molding, shelves, and painting/picture frames (Bornehag et al., 2004, 2005; Hsu et al., 2012; Langer et al., 2010; Kolarik et al., 2008a, 2008b). Most other studies have collected dust from either the floor (Abb et al., 2009; Nagorka et al., 2005; Fromme et al., 2004; Clausen et al., 2002; Kang et al., 2012) or multiple surfaces including the floor (Guo et al., 2011; Becker et al., 2004, 2002; Rudel et al., 2003; Oie et al., 1997). If it is not common to remove shoes at the entrance, dust inside the house may include more substances from outside (Lioy et al., 2002). Only the present study and one previous study have collected both floor and multi-surface dust (Kanazawa et al., 2010). In the study by Kanazawa et al. (2010), only 41 dwellings were examined and they were limited to single-family houses built within the past six years. The present study included various kinds of dwellings, with characteristics different from those included in Kanazawa’s study. In particular, the building structure and construction periods differed, with wooden structured house constituting 46.1% in this study and 79.8% in Kanazawa’s study, and median building age being 10.5 years (range, 0–45.0 years) and 4.8 years (range, 3.2–8.3 years), respectively. According to the Japan Plasticizer Industry Association and Ministry of Economy, Trade, and Industry (2012), a total of 278,896 tons of phthalates were produced in 2008, and 211,465 tons in 2011. Since the annual amount of phthalate production and usage has decreased in Japan, different building periods may be associated with different usage of phthalates for interior materials. In addition, the age of inhabitants in Kanazawa's study differed from that in the present study (mean ± SD: 35.2 ± 21.9 and 26.5 ± 16.7 years, respectively). The inhabitants in the present study were younger than in Kanazawa's study because elementary school children and their parents participated in our study. Generally, dwellings with young children have many plastic materials, such as toys, containers, and shelves; therefore, the dwelling and inhabitant characteristics that differed between this study and Kanazawa's study may have contributed to the different levels of phthalates observed.

This study has several limitations. Firstly, environmental measurements were conducted only once. Seasonal and environmental factors affect the quantity and composition of house dust (Mercier et al., 2011). However, we consciously used the same sampling season from September to November in both 2009 and 2010. Secondly, our study only included dwellings where elementary school children were living. Generally, dwellings with and without young children differ in regards to the numbers of toys and kinds of furniture. Therefore the phthalate levels in dwellings with children might be different from those in dwellings without children. However, the results of the associations between interior materials and levels of phthalates are likely applicable to other situations. Thirdly, since we collected multi-surface dust from many kinds of products at the same time, it was not possible to examine detailed associations between phthalates levels in multi-surface dust and types of surface materials. Therefore, further studies are needed to clarify this hypothesis. Fourthly, although we conducted multivariate regression analyses, the sample size of this study may not have been sufficient for some of these analyses. However, even with a small sample size, our results showed a strong relationship between levels of DEHP and DiBP in floor dust and PVC floors and compressed wooden floors, respectively. Lastly, floor and wall materials were inspected only by observation; therefore, it is possible that some materials were misclassified despite using a pre-established checklist and well-trained inspectors.

In conclusion, this is the first field study to separately evaluate the associations between building characteristics and indoor phthalate levels in both floor and multi-surface dust. The DEHP levels in floor dust were higher in dwellings that had PVC flooring in the main living area, and the DiBP levels in floor dust were increased in dwellings with compressed wooden flooring. Our findings suggest that, in Japanese dwellings, attention should be paid to not only DEHP but also DiBP, which is reportedly an environmental pollutant with allergy adjuvant effects in experimental studies. Furthermore, we found extremely high DEHP levels in multi-surface dust compared with the levels reported in other studies; therefore, especially in Japan, adverse effects of DEHP also should be considered. Phthalate levels were higher in multi-surface dust than in floor dust; thus, when using the house dust as an exposure assessment, it is very important to note where samples were collected from. Overall, this report provides information on how phthalates inside Japanese dwellings are associated with house dust, which can be used in establishing public health provisions.

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Table 1
Comparison between the present home environment survey and the 2008 questionnaire survey.

	Questionnaire survey (n = 4408)		Home environment survey (n = 128)	
Type of dwelling, n (%)				
Single-family house	1868	(43.4)	60	(46.9)
Multi-family house	2355	(54.7)	68	(53.1)
Building structure, n (%)				
Wooden	2200	(51.4)	58	(45.3)
Reinforced concrete	2043	(47.7)	69	(53.9)
Renovation, n (%)	562	(13.5)	31	(24.2)
Age of building, median years (range)	10.5	(0–45)	12	(0–77)
Dampness index (0-4), mean (SD)[a]	1.0	(1.0)	2.1	(1.2)
Condensation, n (%)	2253	(52.4)	92	(71.9)
Visible mold, n (%)	1514	(35.2)	98	(76.6)
Moldy odor, n (%)	228	(5.3)	19	(14.8)
Water leakage, n (%)	485	(11.3)	28	(21.9)
Household income per year, n (%)				
< 3 million yen	Not surveyed		6	(4.7)
3 – 5 million yen			25	(19.5)
5 – 8 million yen			50	(39.1)
>= 8 million yen			28	(21.9)
Unknown			19	(14.8)

[a]The index of the sum of dampness related signs, including condensation, visible mold, moldy odor, and water leakage.

Table 2
Characteristics of buildings in this study (n = 128).

		n (%)
Floor materials in main living area		
	PVC floor	11 (8.6)
	Compressed wooden floor	92 (71.9)
	Wall-to-wall carpet	11 (8.6)
	Tatami/tiles/natural wooden floor	14 (10.9)
PVC wallpaper in main living area		
	Yes	113 (88.3)
	No	15 (11.7)
PVC ceiling in main living area		
	Yes	110 (85.9)
	No	18 (14.1)
Number of PVC interior materials (floor, wall, and ceiling)		
	0	11 (8.6)
	1	6 (4.7)
	2	105 (82.0)
	3	6 (4.7)
Type of dwelling		
	Single-family house	60 (46.9)
	Multi-family apartment	68 (53.1)
Building structure		
	Wooden	58 (45.3)
	Reinforced concrete	69 (53.9)

	Mean (SD)
Age of building (years)	13.3 (10.6)
Floor area (m^2)	15.8 (5.3)
Height of ceiling (cm)	253 (47.8)
Frequency of cleaning living room (times/week)	3.9 (2.1)
Temperature (°C)	21.1 (2.0)
Humidity (%)	54.8 (8.7)

Table 3
Phthalate distribution in dust (μg/g dust; n = 128).

	LOD	Floor					Multi-surface						
		Min	Med	(25%, 75%)	Max	Detection (%)	Min	Med	(25%, 75%)	Max	Detection (%)	P^a	ρ^b
DMP	0.5	<LOD	<LOD	(<LOD, <LOD)	4.6	5.5	<LOD	<LOD	(<LOD, <LOD)	4.15	5.47		
DEP	0.5	<LOD	<LOD	(<LOD, <LOD)	58.7	16.4	<LOD	<LOD	(<LOD, <LOD)	9.76	11.7		
DiBP	0.5	<LOD	3.1	(1.5, 6.1)	97.4	93.0	0.6	2.5	(1.8, 3.60)	26.6	100	0.009	0.293**
DnBP	2.0	<LOD	16.6	(7.5, 32.4)	1670	95.3	<LOD	34.0	(17.2, 75.2)	1380	97.7	<0.001	0.206*
BBzP	1.0	<LOD	2.0	(<LOD, 5.4)	139	68.0	<LOD	3.9	(1.76, 10.5)	267	85.2	<0.001	0.263**
DEHP	1.0	213	1110	(786, 1740)	7090	100	<LOD	2290	(1140, 4460)	44000	99.2	<0.001	0.097
DINP	2.0	11.9	139	(66.1, 276)	2100	100	18.0	203	(99.7, 443)	15500	100	<0.001	0.258**
DEHA	4.0	<LOD	8.0	(4.6, 13.6)	1100	82.8	<LOD	25.9	(13.7, 42.2)	1670	88.3	<0.001	0.233**
BHT	1.0	<LOD	<LOD	(<LOD, 1.1)	5.3	30.5	<LOD	1.2	(<LOD, 2.1)	30.8	60.9	<0.001	0.265**

[a]Significant differences between floor and multi-surface dust were analyzed by Wilcoxon matched rank test.
[b]Spearman's rank correlation test between floor and multi-surface dust.
*$P < 0.05$, **$P < 0.01$.
LOD: Limit of detection; Med: Median.

Table 4

Association between dwelling environment and concentrations of phthalates in floor dust and multi-surface dust (μg/g dust; n = 128).

Floor dust	n	DiBP Med. (25%, 75%)	DnBP Med. (25%, 75%)	BBzP Med. (25%, 75%)	DEHP Med. (25%, 75%)	DINP Med. (25%, 75%)	DEHA Med. (25%, 75%)	BHT Med. (25%, 75%)
Floor materials								
PVC floor	11	**1.8 (2.5, 6.7)****	9.5 (18.1, 38.1)	0.5 (1.2, 5.1)	**2190 (3210, 4530)****	34.2 (139, 286.)	2.0 (4.3, 8.6)	0.5 (0.5, 0.5)
Compressed wooden floor	92	**3.6 (1.8, 6.4)**	14.8 (7.8, 33.3)	1.9 (0.5, 6.2)	**1060 (773, 1620)**	148 (70.3, 324)	7.8 (4.9, 14.2)	0.5 (0.5, 1.1)
Wall-to-wall carpet	11	**1.1 (0.6, 1.8)**	10.6 (5.1, 25.7)	2.2 (0.5, 5.5)	**1120 (805, 1460)**	127 (78.0, 188)	11.2 (7.9, 19.2)	0.5 (0.5, 1.9)
Tatami/tiles/natural wooden floor	14	**2.8 (0.8, 4.3)**	28.0 (6.8, 54.1)	1.6 (0.5, 3.2)	**916 (472, 1720)**	110 (49.8, 271)	8.7 (2.0, 13.8)	0.5 (0.5, 1.6)
PVC wall paper								
No	15	2.1 (0.8, 4.5)	26.2 (5.9, 65.7)	2.0 (0.5, 4.3)	1180 (883, 1750)	144 (88.5, 381)	7.9 (2.0, 15.4)	0.5 (0.5, 1.3)
Yes	113	3.1 (1.6, 6.1)	15.1 (7.6, 30.9)	1.8 (0.5, 5.7)	1110 (777, 1740)	138 (60.4, 271)	8.1 (4.7, 13.6)	0.5 (0.5, 1.1)
PVC ceiling								
No	18	3.7 (1.4, 5.9)	**29.5 (16.7, 69.3)****	2.0 (0.5, 3.9)	1440 (976, 3720)	142 (90.1, 376.5)	7.9 (2.0, 12.8)	0.5 (0.5, 0.7)
Yes	110	2.9 (1.5, 6.1)	**14.4 (6.9, 30.8)**	1.8 (0.5, 5.9)	1070 (747, 1660)	137 (64.7, 271)	8.0 (4.6, 13.7)	0.5 (0.5, 1.2)
Number of PVC interior materials (floor, wall, and ceiling)								
0	11	3.5 (0.9, 5.6)	32.7 (19.6, 65.7)	2.0 (1.2, 3.2)	1040 (883, 1670)	171 (117, 393)	8.0 (2.0, 17.7)	0.5 (0.5, 1.2)
1	6	1.3 (0.3, 2.7)	15.8 (5.4, 225)	3.4 (0.5, 11.2)	1490 (938, 5050)	82.2 (45.1, 217)	7.1 (2.0, 9.7)	0.5 (0.5, 2.3)
2	105	3.4 (1.6, 6.2)	14.7 (7.3, 30.9)	1.8 (0.5, 6.0)	1070 (746, 1660)	136 (62.7, 261)	8.3 (4.8, 13.8)	0.5 (0.5, 1.1)
3	6	2.3 (1.6, 3.6)	20.1 (7.4, 26.9)	2.4 (1.0, 6.7)	2260 (1370, 3210)	229 (53.4, 1090)	4.0 (2.0, 14.0)	0.5 (0.5, 1.3)
Type of dwelling								
Single-family house	68	2.5 (1.2, 5.9)	18.2 (7.9, 35.7)	2.3 (0.1, 5.4)	1210 (805, 2040)	137 (69.6, 224)	7.0 (4.3, 12.7)	0.5 (0.5, 1.3)
Multi-family apartment	60	3.2 (1.9, 6.3)	14.4 (6.8, 32.4)	1.7 (0.5, 6.4)	1060 (727, 1660)	140 (61.1, 403)	8.3 (5.1, 15.2)	0.5 (0.5, 1.1)
Building structure								
Wooden	59	2.5 (1.2, 5.8)	18.2 (7.9, 37.5)	2.2 (0.5, 5.9)	1210 (802, 2090)	138 (80.6, 219)	6.8 (4.2, 12.6)	0.5 (0.5, 1.2)
Reinforced concrete	69	3.4 (1.9, 6.6)	14.1 (6.9, 32.2)	1.8 (0.5, 4.9)	1070 (734, 1640)	138 (57.2, 387)	8.4 (5.6, 15.1)	0.5 (0.5, 1.1)
Dampness Index (0-5)								
0	17	3.0 (1.6, 6.5)	27.4 (7.4, 42.7)	2.0 (1.2, 5.1)	1060 (582, 1670)	107 (48.9, 401)	8.8 (6.0, 27.4)	0.5 (0.5, 1.1)
1	22	2.9 (1.9, 6.0)	10.7 (5.7, 23.6)	1.3 (0.5, 6.5)	949 (723, 1200)	115 (53.0, 148)	6.7 (2.0, 11.8)	0.5 (0.5, 0.5)
2	37	3.1 (1.5, 5.7)	16.1 (7.3, 31.2)	2.0 (0.5, 4.5)	1150 (779, 1990)	189 (88.1, 436)	9.4 (6.1, 15.7)	0.5 (0.5, 1.3)
3	38	2.0 (1.1, 5.6)	17.9 (7.7, 28.6)	2.0 (0.5, 5.2)	1180 (744, 2060)	148 (60.2, 255)	7.8 (4.7, 10.9)	0.5 (0.5, 1.0)
4	11	4.3 (1.7, 11.7)	61.6 (7.1, 159)	1.3 (0.5, 9.5)	1610 (871, 2060)	146 (48.4, 277)	6.5 (2.0, 8.6)	0.5 (0.5, 2.4)
5	3	5.9 (5.6, 11.3)	16.1 (10.1, 21.1)	3.2 (0.5, 6.3)	1440 (663, 3210)	81.4 (22.6, 200)	4.5 (2.0, 44.0)	0.5 (0.5, 1.6)
Annual household income (yen/year)								
< 3 million	6	4.9 (2.0, 5.9)	30.1 (8.5, 126.8)	0.8 (0.5, 4.9)	2500 (979, 3380)	161 (28.1, 1010)	**3.1 (2.0, 6.4)**	0.5 (0.5, 2.4)
3 – < 5 million	25	2.7 (1.4, 5.7)	14.8 (7.8, 22.8)	0.5 (0.5, 3.7)	1120 (658, 2090)	132 (50.2, 224)	**7.8 (2.8, 12.3)**	0.5 (0.5, 1.0)
5 – < 8 million	50	3.7 (1.6, 6.2)	18.4 (6.9, 35.0)	1.7 (0.5, 6.0)	1060 (801, 1660)	189 (76.0, 378)	**8.9 (6.1, 16.3)**	0.5 (0.5, 1.5)
>= 8 million	28	3.7 (1.4, 9.1)	19.8 (8.0, 35.8)	2.7 (0.6, 6.5)	1190 (884, 2060)	129 (83.6, 267)	**9.0 (4.4, 17.5)**	0.5 (0.5, 1.3)
Unknown	19	1.8 (1.4, 2.9)	11.9 (6.7, 61.6)	2.5 (0.5, 6.3)	980 (719, 1570)	121 (83.9, 172)	**5.9 (2.0, 9.1)***	0.5 (0.5, 0.5)
		ρ	ρ	ρ	ρ	ρ	ρ	ρ
Age of building (yr)		−0.044	**0.241****	0.016	**0.235****	0.016	0.026	0.079
Height of ceiling (cm)		0.007	−0.112	0.047	−0.155	0.036	0.02	−0.080
Frequency of cleaning living room (times/week)		**0.197***	0.014	−0.027	−0.133	−0.107	−0.025	−0.035

Multi-surface dust	n	DiBP Med. (25%, 75%)	DnBP Med. (25%, 75%)	BBzP Med. (25%, 75%)	DEHP Med. (25%, 75%)	DINP Med. (25%, 75%)	DEHA Med. (25%, 75%)	BHT Med. (25%, 75%)
Floor materials								
PVC floor	11	2.2 (2.4, 5.9)	29.9 (36.7, 66.6)	0.5 (2.4, 5.1)	1840 (3730, 5030)	72.4 (176, 348)	23.5 (32.6, 41.9)	0.5 (1.6, 3.9)
Compressed wooden floor	92	2.6 (1.7, 3.6)	29.7 (14.5, 59.1)	3.9 (1.8, 9.7)	2270 (1220, 4170)	228 (120, 543)	24.3 (13.5, 39.4)	1.2 (0.5, 1.9)
Wall-to-wall carpet	11	2.1 (1.6, 4.3)	53.3 (27.2, 229)	10.4 (1.8, 14.8)	2570 (824, 3510)	203 (127, 276)	31.3 (17.8, 51.7)	1.7 (0.5, 4.0)
Tatami/tiles/natural wooden floor	14	2.5 (1.9, 3.1)	37.0 (19.9, 89.3)	5.7 (1.5, 20.5)	1140 (739, 5950)	105 (72.3, 236)	36.7 (10.9, 192)	0.5 (0.5, 1.7)
PVC wall paper								
No	15	2.1 (1.5, 2.5)	76.6 (32.1, 126)	5.1 (1.8, 19.6)	**888 (614, 2880)***	**117 (76.8, 157)****	26.1 (19.8, 51.7)	1.2 (0.5, 1.8)
Yes	113	2.6 (1.8, 3.7)	30 (16.9, 58.2)	3.8 (1.7, 10.0)	**2400 (1300, 5030)**	**241 (105, 522)**	25.6 (13.6, 41.7)	1.2 (0.5, 2.1)
PVC ceiling								
No	18	2.2 (1.8, 3.0)	**55.7 (32.0, 136)***	5.3 (2.5, 16.0)	1540 (739, 4010)	**130 (68.3, 159)****	32.0 (22.8, 49.9)	1.6 (0.5, 3.4)
Yes	110	2.6 (1.8, 3.6)	**30.0 (16.5, 57.3)**	3.8 (1.6, 9.9)	2450 (1250, 5030)	**244 (108, 541)**	25.5 (13.6, 40.8)	1.2 (0.5, 2.0)
Number of PVC interior materials (floor, wall, and ceiling)								
0	11	1.9 (1.5, 2.4)	76.6 (23.9, 126)	11.2 (4.2, 23.7)	**814 (594, 2280)***	**133 (71.5, 157)****	26.1 (19.8, 67.8)	0.5 (0.5, 1.8)
1	6	3.2 (2.2, 5.1)	75.9 (31.5, 174)	2.6 (0.5, 6.6)	**2450 (1790, 3040)**	**122 (91.4, 172)**	32.4 (18.9, 44.6)	1.6 (1.2, 9.7)
2	105	2.6 (1.8, 3.7)	31.2 (16.5, 58.2)	3.8 (1.6, 10.3)	**2400 (1190, 5030)**	**241 (105, 542)**	26.1 (13.5, 41.7)	1.2 (0.5, 2.2)
3	6	2.3 (2.1, 5.7)	29.9 (20.7, 72.8)	3.2 (1.4, 5.5)	**3550 (2260, 5450)**	**329 (92.9, 592)**	25.0 (18.1, 43.7)	0.8 (0.5, 1.3)
Type of dwelling								
Single-family house	60	2.6 (1.9, 3.8)	34.9 (21.6, 65.0)	3.8 (1.8, 9.6)	**2800 (1600, 5570)***	235 (114, 531)	24.5 (13.8, 39.5)	**1.4 (0.5, 2.7)***
Multi-family apartment	68	2.4 (1.7, 3.6)	32.3 (13.1, 77.8)	4.0 (1.6, 11.0)	**1990 (876, 3330)**	188 (88.2, 364)	28.3 (13.5, 56.8)	**1.1 (0.5, 1.8)**
Building structure								
Wooden	59	2.5 (1.8, 3.4)	36.6 (21.5, 66.6)	3.9 (1.8, 10.4)	**2640 (1590, 5660)***	163 (83.9, 361)	27.5 (13.4, 47.6)	1.2 (0.5, 1.9)
Reinforced concrete	69	2.5 (1.7, 3.7)	30.0 (13.2, 77.4)	4.1 (1.6, 12.1)	**1920 (880, 3460)**	252 (129, 544)	24.5 (13.8, 41.5)	1.3 (0.5, 2.6)
Dampness Index (0-5)								
0	17	3.1 (1.8, 3.6)	**14.2 (10.8, 31.9)****	3.4 (1.3, 9.3)	**1150 (724, 3080)***	204 (77.1, 555)	27.1 (11.0, 56.4)	**1.1 (0.5, 1.3)***
1	22	2.4 (1.7, 3.1)	**29.2 (13.9, 85.9)**	4.6 (1.6, 10.9)	**1990 (872, 4360)**	273 (107, 424)	29.9 (12.5, 50.6)	**0.8 (0.5, 1.8)**
2	37	2.3 (1.7, 3.1)	**33.3 (19.0, 65.7)**	3.8 (1.9, 11.1)	**2450 (1200, 5150)**	161 (125, 364)	24.1 (13.7, 41.3)	**1.2 (0.5, 2.0)**
3	38	2.6 (1.9, 3.6)	**37.0 (19.8, 83.8)**	4.3 (1.3, 14.9)	**2520 (1700, 5050)**	257 (107, 936)	25.6 (13.9, 41.5)	**1.2 (0.5, 2.4)**
4	11	3.4 (2.6, 7.7)	**54.5 (36.7, 414.9)**	3.4 (2.1, 4.2)	**2280 (1120, 5030)**	141 (72.4, 174)	33.3 (24.5, 42.2)	**1.8 (1.5, 3.6)**
5	3	4.1 (2.9, 5.9)	**32.6 (3.6, 100.4)**	7.1 (3.4, 9.8)	**4180 (1360, 6410)**	72.9 (48.6, 89.1)	21.9 (2.0, 37.8)	**4.1 (1.9, 7.0)**
Annual household income (yen/year)								
< 3 million	6	3.6 (2.8, 8.2)	46.4 (28.0, 229)	3.7 (3.1, 10.1)	2830 (1750, 4250)	148 (66.4, 295)	23.2 (2.0, 40.6)	1.7 (1.2, 3.8)
3 – < 5 million	25	2.9 (2, 3.6)	27.4 (16.5, 48.5)	3.2 (1.4, 7.5)	2450 (1780, 5490)	203 (122, 340)	23.1 (13.7, 41.7)	0.5 (0.5, 1.8)
5 – < 8 million	50	2.5 (1.6, 3.7)	32.2 (15.4, 83.8)	3.8 (1.4, 9.2)	2520 (1190, 6340)	218 (97.0, 565)	27.6 (13.7, 43.2)	1.1 (0.5, 2.2)
>= 8 million	28	2.5 (1.9, 3.3)	40.1 (21.2, 94)	4.5 (2.0, 18.5)	1820 (815, 4240)	203 (78.6, 981)	31.1 (13.0, 63.8)	1.3 (0.5, 1.9)
Unknown	19	2.3 (1.7, 3.2)	29.9 (17.1, 53.3)	3.9 (1.8, 10.6)	1840 (814, 2640)	204 (98.9, 379)	25.5 (18.7, 41.7)	1.4 (0.5, 2.5)
		ρ	ρ	ρ	ρ	ρ	ρ	ρ
Age of building (yr)		0.139	**0.460****	**0.188***	**0.180***	−0.024	0.106	**0.227****
Height of ceiling (cm)		−0.123	**−0.201***	−0.153	**−0.253****	0.083	−0.093	**−0.179***
Frequency of cleaning living room (times/week)		0.07	−0.154	0.05	0.069	−0.009	−0.014	0.027

Statistical significance was calculated using the nonparametric Mann-Whitney U test for the two categorical variables. Associations between phthalate concentrations and flooring materials and household income were

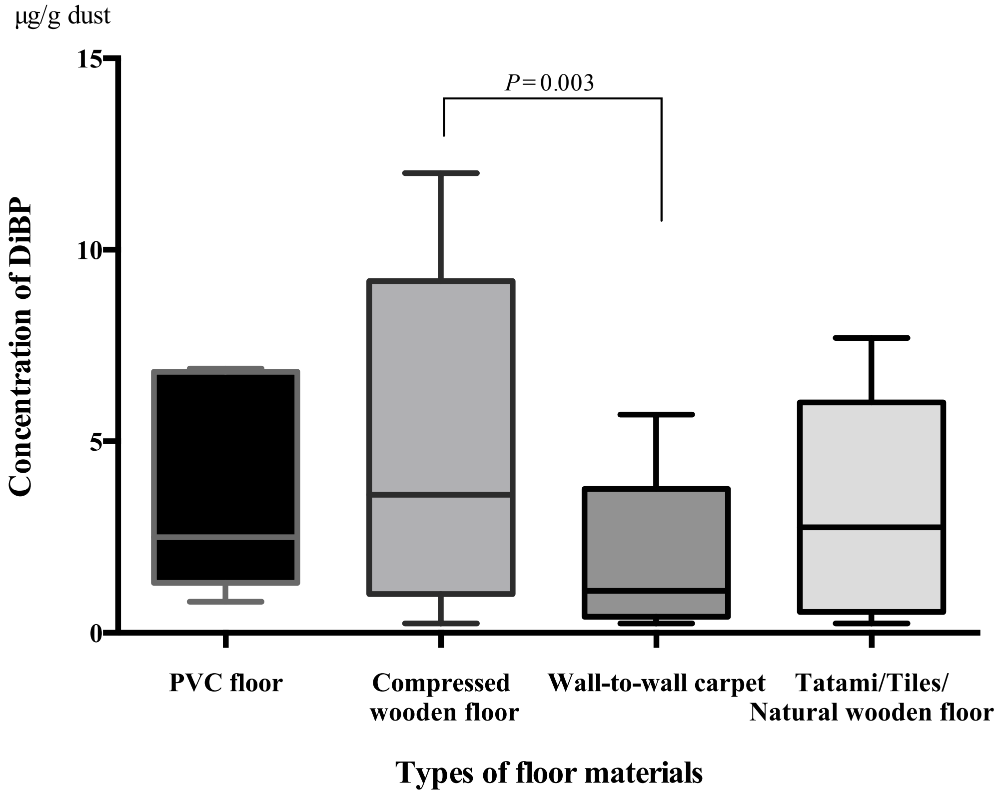
Table 5
Comparison of phthalate levels in house dust in different studies.

Sampling place and Study	Country	n	Median (µg/g dust)			
			DEHP	BBzP	DnBP	DINP
Floor dust						
Present study	Japan (Sapporo)	128	1107	2	17	139
Kang et al. 2012	China	23	1190	5	77	-
Guo et al. 2011	USA	33	304	21	20	-
Kanazawa et al. 2010	Japan (Sapporo)	41	880	4	19	126
Abb et al. 2009	Germany	30	604	15	87	129
Nagorka et al. 2005	Germany	278	480	13	29	80
Fromme et al. 2004	Germany	30	700	30	60	-
Clausen et al. 2002	Denmark	23	858	-	-	-
Multi-surface dust excluding the floor[b]						
Present study	Japan (Sapporo)	128	2293	4	34	203
Hsu et al. 2012	Taiwan	76–92	753	1	20 (DBP)	-
Kanazawa et al. 2010	Japan (Sapporo)	41	1200	2	22	116
Langer et al. 2010	Denmark	497	210	3.7	15	-
Kolalik et al. 2008	Bulgaria	184	990	330	9850	-
Bornehag et al. 2004	Sweden	346	770	135	150	40
Multi-surface dust including the floor[c]						
Guo et al. 2011	China	75	228	0.2	20	-
Becker et al. 2004	Germany	252	515	-	-	-
Rudel et al. 2003	USA	120	340	45	20	-
Becker et al. 2002	Germany	199	416	15	42	
Ole et al. 1997	Norway	38	640[a]	11[a]	-	-

[a]Mean concentration.
[b]Dust was collected from multiple surfaces excluding the floor surface.
[c]Dust was collected from multiple surfaces including the floor surface.

Fig.1.

a.


μg/g dust
15
10
5
0
Concentration of DiBP
P = 0.003
PVC floor
Compressed wooden floor
Wall-to-wall carpet
Tatami/Tiles/ Natural wooden floor
Types of floor materials

b.

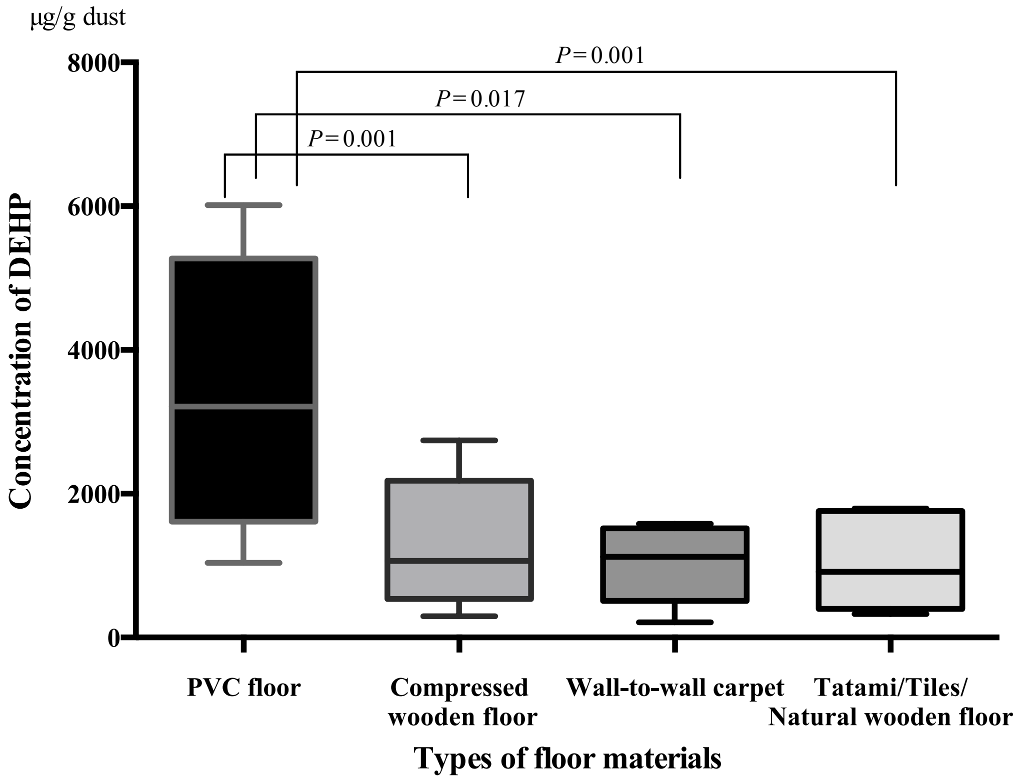
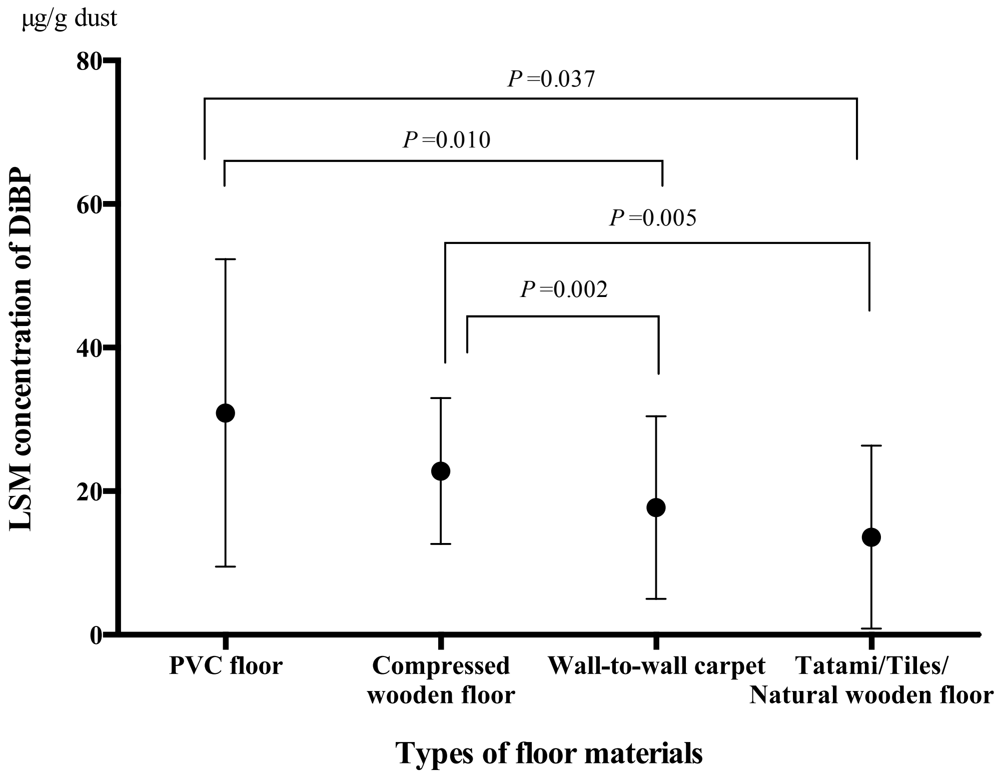

μg/g dust
8000
6000
4000
2000
0
Concentration of DEHP
P = 0.001
P = 0.017
P = 0.001
PVC floor
Compressed wooden floor
Wall-to-wall carpet
Tatami/Tiles/ Natural wooden floor
Types of floor materials

Fig. 2

a.


μg/g dust
80
60
40
20
0
P =0.037
P =0.010
P =0.005
P =0.002
LSM concentration of DiBP
PVC floor
Compressed wooden floor
Wall-to-wall carpet
Tatami/Tiles/ Natural wooden floor
Types of floor materials

b.

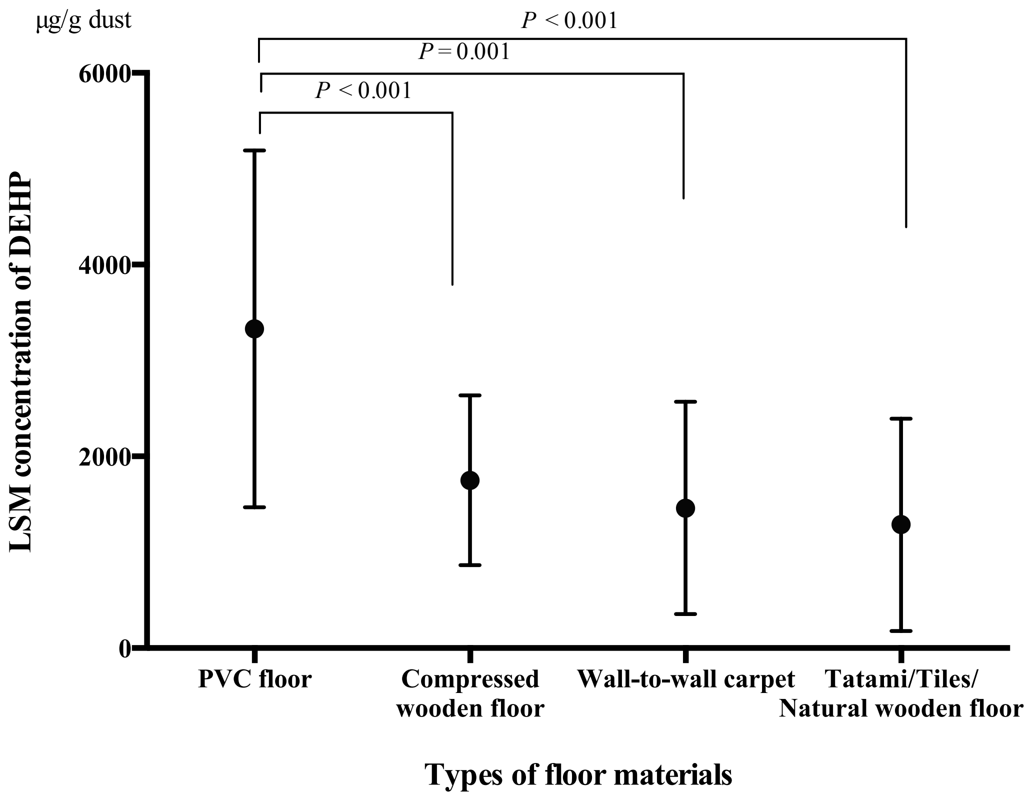
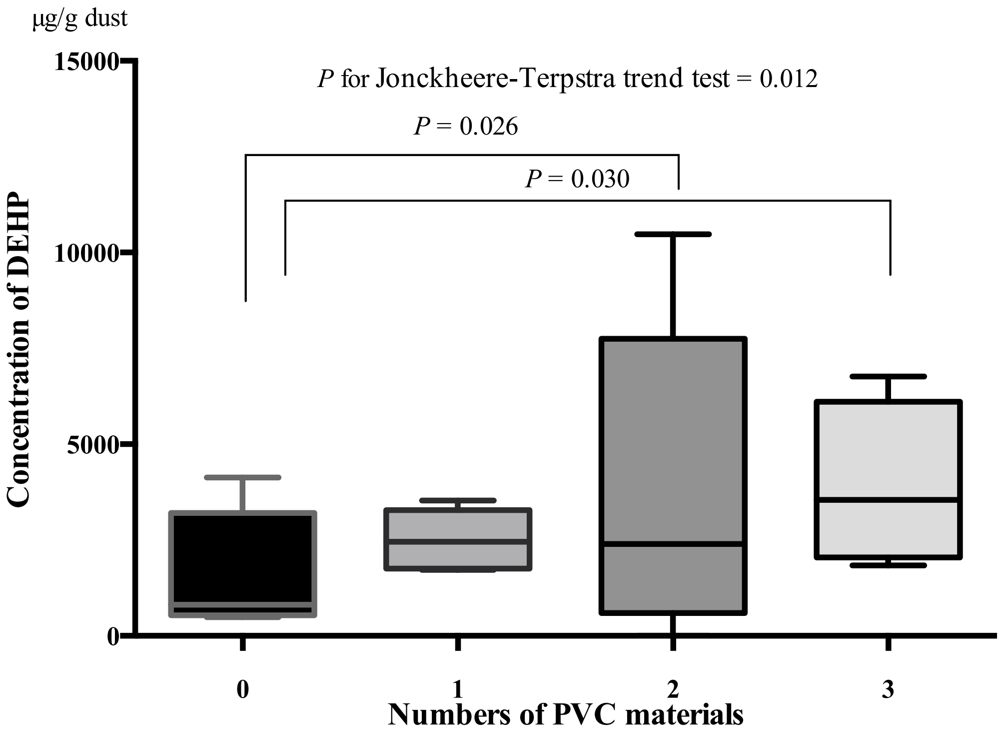

μg/g dust
6000
4000
2000
0
P < 0.001
P = 0.001
P < 0.001
LSM concentration of DEHP
PVC floor
Compressed wooden floor
Wall-to-wall carpet
Tatami/Tiles/ Natural wooden floor
Types of floor materials

Fig. 3

a.


μg/g dust
15000
10000
5000
0
P for Jonckheere-Terpstra trend test = 0.012
P = 0.026
P = 0.030
Concentration of DEHP
0
1
2
3
Numbers of PVC materials

b.

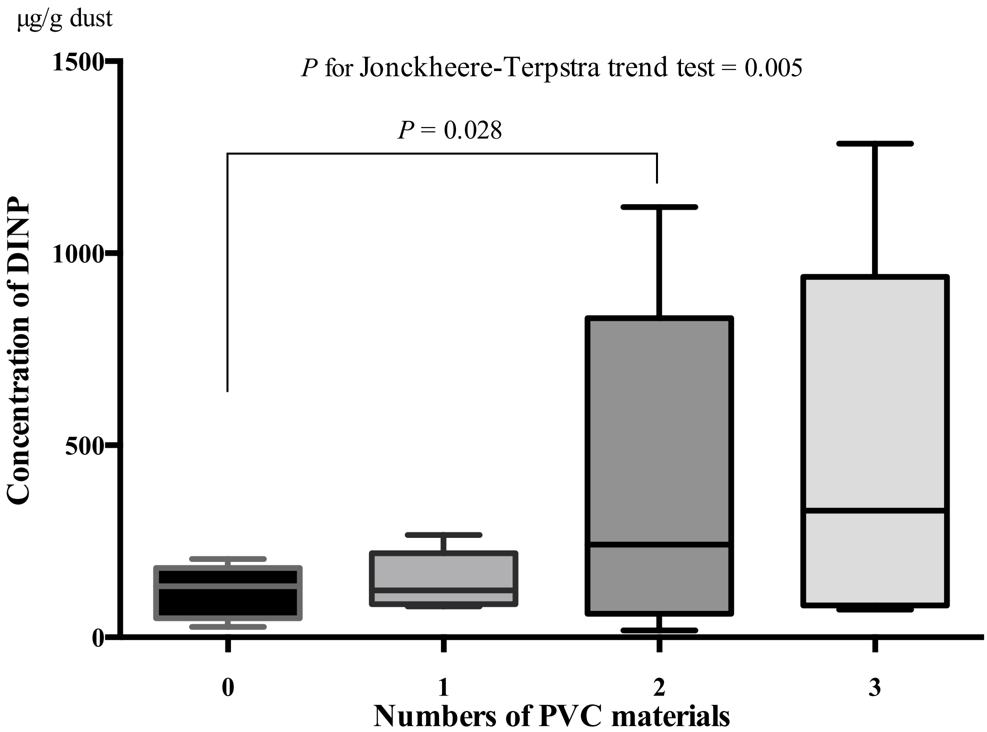
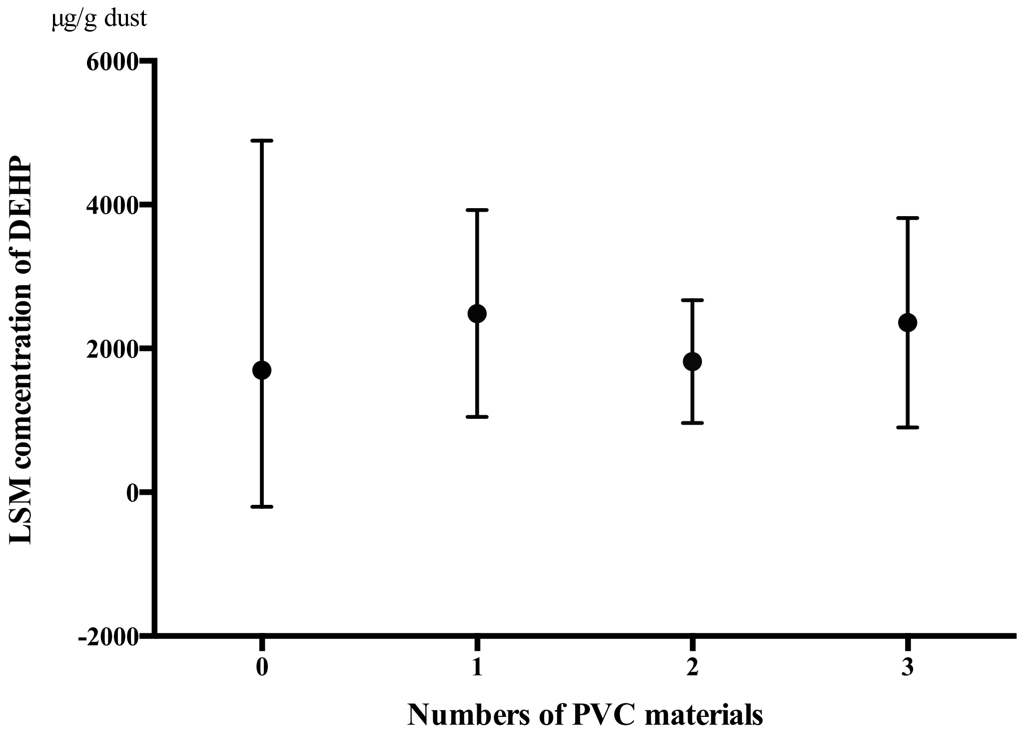

μg/g dust
1500
1000
500
0
P for Jonckheere-Terpstra trend test = 0.005
P = 0.028
Concentration of DINP
0
1
2
3
Numbers of PVC materials

Fig. 4

a.


μg/g dust
6000
4000
2000
0
-2000
LSM comcentration of DEHP
0
1
2
3
Numbers of PVC materials

b.

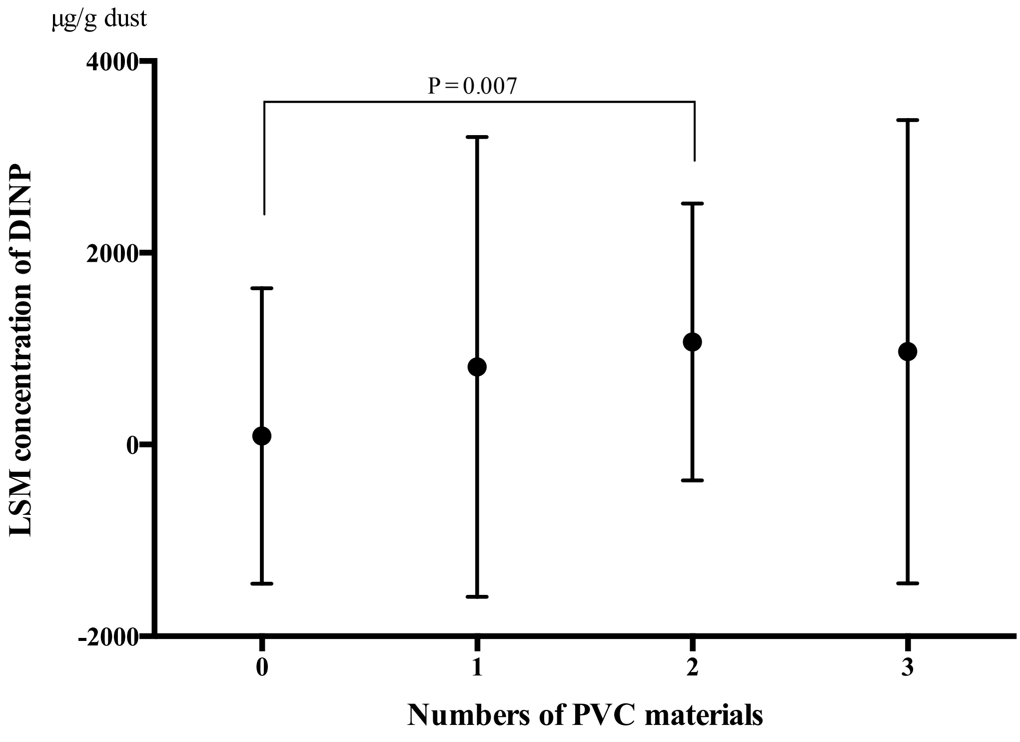

μg/g dust
4000
2000
0
-2000
P = 0.007
LSM concentration of DINP
0
1
2
3
Numbers of PVC materials

Figure legends

Fig. 1. Concentrations of DiBP (a) and DEHP (b) in floor dust from different flooring materials. Box plots show 25% and 75%, with a bold line indicating median concentration, and error bars showing the concentration range. The comparisons among flooring materials were analyzed using the Kruskal-Wallis test, and *P* values were adjusted using Bonferroni's correction. *P* values of less than *P* =0.05 are shown in the figures. Statistical significance of the *P* value was $P < 0.017$ based on Bonferroni's correction.

Fig. 2. Multivariate regression analyses of concentrations of DiBP (a) and DEHP (b) in floor dust and different flooring materials. The associations between phthalates levels and each flooring materials were analyzed using multiple comparisons analyses. Levels of DiBP and DEHP were adjusted for dampness index and household income. Plots show the levels of least square mean (LSM) of DiBP (a) and DEHP (b), and error bars showing the 95% confidencial intervals (95% CI). *P* values of less than *P* =0.05 are shown in the figures. After multiple comparison, statistically significance of the *P* value was $P < 0.017$ based on Bonferroni's correction.

Fig. 3. The correlations between the concentrations of DEHP (a) and DINP (b) in multi-surface dust and the number of PVC interior materials. Box plots show 25% and 75% concentrations, with a bold line indicating median concentration, and error bars showing the concentration range. The correlation between the numbers of PVC materials and phthalate levels were analyzed using the Jonckheere-Terpstra trend test and multiple comparison tests. Multiple comparison tests were conducted to set up "Numbers of PVC materials = 0" as a control. *P* values of less than *P* =0.05 are shown in the figures. After multiple comparison, statistically significance of the *P* value was $P < 0.017$ based on Bonferroni's correction.

Fig. 4. Multivariate regression analyses of concentrations of DEHP (a) and DINP (b) in multi-surface dust and the number of PVC interior materials. Associations between phthalates levels and number of PVC interior materials were analyzed using multiple comparisons analyses. Levels of DEHP and DINP were adjusted for dampness index and household income. Plots show levels of least square mean (LSM) of DEHP (a) and DINP (b), and error bars showing the 95% confidencial intervals (95% CI). Multiple comparison test were conducted to set up “Numbers of PVC materials = 0” as the control. *P* values of less than P =0.05 are shown in the figures. After multiple comparison, statistically significance of the *P* value was $P < 0.017$ based on Bonferroni’s correction.