



Research article

Impact of mold growth on di(2-ethylhexyl) phthalate emission from moist wallpaper

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ARTICLE INFO

Keywords:

Wallpaper
Di(2-ethylhexyl) phthalate
Moist
Fungi
DEHP emission
Indoor

ABSTRACT

Flood damage can increase indoor concentrations of di(2-ethylhexyl) phthalate (DEHP) and molds in households with wallpaper. Wallpaper water content can affect its DEHP emission into indoor environments; however, the influence of mold growth on this DEHP emission remains unclear. Here, we evaluated whether mold growth affects DEHP emission from moist wallpaper (moist WP). Experiments were conducted in glass chambers with wallpaper containing 12.7% (w/w) DEHP and a dust tray sample system at approximately 28 °C and 100% relative humidity (RH). The experimental groups were (1) moist WP, (2) moist WP + *Aspergillus versicolor* (AV), (3) moist WP + *Cladosporium cladosporioides*, (4) moist WP + *Penicillium chrysogenum*, and (5) moist WP + mold mixture. Mold growth on the wallpaper and DEHP emission into air and onto dust were analyzed at nine time-points over 30 days. Initially, the moist WP group emitted relatively high concentrations of DEHP into air, but after at least 8 days, the concentration of DEHP emitted by the mold-added groups exceeded that of the moist WP group. DEHP emission onto dust, especially from the moist WP group, increased considerably at day 15. During the experimental period, the moist WP (13.63 ± 4.67 µg) and moist WP + AV (13.93 ± 0.49 µg) groups emitted higher cumulative amounts of DEHP onto dust. During the 30-day experimental period, obvious mold growth occurred over days 15–30. Moreover, the moist WP group exhibited relatively higher and lower cumulative DEHP emission into air than the mold-added groups during days 2–10 (2.71 vs. 1.94–2.94 µg) and 15–30 (1.16 vs. 1.61–2.12), respectively; a contrasting trend was observed for cumulative DEHP emission onto dust. In conclusion, mold growth affects DEHP emission from water-damaged wallpaper, and the removal or cleaning of wet wallpaper, particularly those with visible mold growth, is critical from a public health perspective.

1. Introduction

Di(2-ethylhexyl) phthalate (DEHP) is a phthalate ester widely used as a plasticizer. According to global market sales, DEHP accounts for 50% of all plasticizers, and DEHP is added to at least 95% of polyvinyl chloride (PVC) products such as containers, toys, raincoats, and building materials (Gevao et al., 2013; Sampath et al., 2017). Building materials, such as PVC flooring and wallpaper, may contain approximately 10% DEHP (w/w) or more (Gong et al., 2014; Lee et al., 2014). DEHP is not chemically bound to the PVC matrix; thus, DEHP emission from products into the environment occurs frequently.

DEHP is distributed in many indoor environmental media, including air, airborne particles, dust, and furniture surfaces (Kashyap and Agarwal 2018; Liu and Zhang 2016). Of all phthalate esters, DEHP demonstrates the highest abundance in indoor air and dust (Huang et al., 2020; Lee et al., 2021; Wang et al., 2021). However, in indoor environments, DEHP is mainly present in the particle phase rather than in the gas phase owing to its low volatility (Chi et al., 2017; Kashyap and Agarwal 2018; Liu and Zhang 2016). Consequently, in households, DEHP concentrations are higher on settled dust than in other media (Bornehag et al., 2005). Moreover, of all phthalate esters found on settled dust indoors, DEHP has the highest concentration (Bergh et al., 2011; Bornehag et al., 2005). A

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Received 2 January 2022; Received in revised form 16 May 2022; Accepted 17 August 2022

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study in Stockholm, Sweden, found that DEHP on settled dust in private homes, daycare centers, and even workplaces accounted for >80% of dust phthalate esters (Bergh et al., 2011).

DEHP emission from PVC products is affected by environmental factors. Hsu et al. (2012) found that DEHP levels were higher in dust collected from houses that had experienced flooding, indicating that moisture content might affect DEHP emission from building materials into air and dust. The higher the moisture content of source materials is, the more extensive is the migration of DEHP within polymers and the higher is its emission into the environment (Kashyap and Agarwal 2018). Moreover, in building materials, DEHP hydrolyzes to 2-ethyl-1-hexanol (2E1H) in the presence of moisture (Yokota et al., 2013). As such, high moisture content enhances the rate of DEHP reduction through hydrolysis. This reduces DEHP levels in the boundary and thereby creates a DEHP concentration gradient toward the boundary (Hsu et al., 2017). In their chamber study, Hsu et al. (2017) further determined that moist wallpaper (moist WP) emits 35.31% more DEHP mass into air, dust, and wipe samples than does dry wallpaper.

Mold affects water-damaged rooms more extensively than those without any water damage (Gravesen et al., 1999; Haas et al., 2007; Walinder et al., 2001). After floods, wet building materials support mold growth (Brandt et al., 2006; Riggs et al., 2008) and the emission of DEHP (Hsu et al., 2012). This is because mold uses organics within building materials, such as wallpaper, as nutrients for its growth, consequently leading to DEHP emission. A study in China reported a significantly positive association between urinary concentrations of DEHP metabolites in pregnant women and moldy walls (Dong et al., 2020). Therefore, we hypothesized that wallpaper with a high water content would favor mold growth, which would increase DEHP emission into the environment. However, no study has demonstrated the association between the mold growth and DEHP emission.

With global climate change, the frequency of extreme weather events, including heavy rainfall, has increased. Every 1 °C increase in local temperature can increase moisture content in the global atmosphere by approximately 7% (Kininmonth 2010). This can lead to more intense precipitation events, resulting in an increased frequency of floods and consequently increasing the possibility of mold growth on moist WP and the subsequent emission of DEHP from wallpaper into the environment. DEHP exposure orally, dermally, and through inhalation is associated with several adverse health outcomes such as those relating to reproductive toxicity (Barakat et al., 2017; Pocar et al., 2017), testicular toxicity (David 2006), neurotoxicity (Wu et al., 2019), cardiotoxicity (Amara et al., 2019; Wang et al., 2019), asthma, and allergies (Wang et al., 2015). This study, therefore, evaluated whether mold growth affects DEHP emission from moist WP into the environment and compared the differences in the effects of different mold species on DEHP emission levels.

2. Materials and methods

2.1. Chamber system and experimental materials

A 5.8-L glass chamber was used to perform our experiments. The major components of the chamber system were temperature and humidity sensors (HOBO temperature logger; U10-003, Onset Computer, Bourne, MA, USA), a temperature controller model (NewLab HT-720, Taiwan), four pieces of wallpaper (three rectangular pieces, each sized 12.5 cm × 60 cm, 5 cm × 44.5 cm, and 9.5 cm × 15.5 cm); one circular piece, with a 13.8-cm diameter), dust platforms (16.3 cm × 10 cm × 10 cm), sorbent tubes for air sampling, nine dust tray sample systems, and a magnet stirrer (SP88857100; Thermo Fisher Scientific, Waltham, MA, USA). Each dust tray sample system (1.1 cm × 1.1 cm × 10.2 cm) contained 1 g of the standard dust ASHRAE 52–76, composed of 72% Arizona road dust (0–80 μm in diameter), 23% carbon black, and 5% fibers. Before the experiments, the standard dust was soaked in n-pentane overnight and then shaken ultrasonically in fresh n-pentane for 30 min;

this was repeated three times. The treated standard dust was dried in a chemical hood to avoid background contamination. A magnet stirrer was placed at the bottom of the chamber to ensure an even distribution of the environmental components inside the chamber during the entire experiment. The chambers were airtight without any ventilation, except at sample collection time-points.

One of the best-selling PVC-coated wallpapers, SKURA 71464, was selected for our study because of its high (12.7% w/w) DEHP concentration (Hsu et al., 2017). Moreover, this wallpaper is coated with PVC on only one side.

The three airborne mold species most commonly found indoors after flooding in Taiwan were selected for the experiments: (1) *Aspergillus versicolor* (BCRC 31895), (2) *Cladosporium cladosporioides* (BCRC 30812), and (3) *Penicillium chrysogenum* (BCRC 30873). These species were grown on dichloran glycerol (DG18) agar at 25 °C for 30 days. Next, the spores were extracted from the medium by using sterile 0.5% Tween (Sigma-Aldrich, St. Louis, MO, USA) and were then diluted to a concentration of $(2.35\text{--}2.42) \times 10^6$ spores/mL. This suspension was then added to the moist WP samples by using a glass dropper; in particular, 1 mL of the diluted spore suspension was added per 25 cm² of the moist WP and spread using a glass spreader to ensure an even distribution. As such, the four wallpaper pieces with the areas of 750, 222.5, 149.5, and 147.5 cm² were exposed to 30, 8.9, 6.0, and 5.9 mL of the diluted spore suspension, respectively.

2.2. Experiment design

To evaluate the influence of mold species on DEHP emission from wallpaper samples under high moisture content, four experimental conditions (varying by the mold species) coupled with a control condition without the addition of mold were examined (Figure S1). In all five chamber systems, the temperature and RH were maintained at approximately 28 °C and 100% RH, respectively (Hsu et al., 2017), based on the annual average indoor temperature and RH after flooding in Taiwan, respectively. After the temperature and RH in the chamber systems became stable, the moist WP samples with and without mold addition were placed inside the chambers, and the experiments were initiated. If RH decreased to lower than 90% ± 10% during the experimental period, 1 mL of sterile ultrapure water was injected into the chamber. The wallpaper samples in the five chambers were denoted as follows:

- (I) Moist WP alone (moist WP) (reference for comparison)
- (II) Moist WP spiked with *A. versicolor* (moist WP-AC)
- (III) Moist WP spiked with *C. cladosporioides* (moist WP-CC)
- (IV) Moist WP spiked with *P. chrysogenum* (moist WP-PC)
- (V) Moist WP spiked with *A. versicolor*, *C. cladosporioides*, and *P. chrysogenum* (moist WP-Mix)

The moisture content of all wallpaper samples ranged from 52.44% to 54.35%. All the experiments were performed in triplicate. The experimental period was 30 days. To monitor the moisture content of wallpapers, a fixed-size piece (5 cm × 5 cm) was cut and analyzed on experiment days 2, 4, 6, 8, 10, 15, 20, 25, and 30. At each time-point, the fixed-size piece of wallpaper was taken out of the chamber for moisture content measurement on an Infrared Moisture Determination Balance (FD-610, Kett, Japan). Next, the wallpaper was humidified to saturation and then placed back in the chamber. The coefficient of variation (CV) of RH and moisture content during the 30 days was noted to be <5%.

2.3. Sample collection

Figure S2 presents the design of the experimental chamber. Air samples were collected using sorbent tubes (Tenax TA OVS; SKC 226-56; SKC, Eighty Four, PA, USA) at a flow rate of 1 L/min (Gillian GilAir-5; Sensidyne, St. Petersburg, FL, USA) for 4 h on days 2, 4, 6, 8, 10, 15,

20, 25, and 30 for DEHP analysis. During air sampling, the chamber was continuously purged with fresh ambient air to ensure the complete collection of the DEHP mass inside the chamber. The fresh air inlet was equipped with an active carbon filter (Honeywell, NC, USA) for the removal of DEHP in ambient air. The air around the chamber was also collected to check for any background contamination with DEHP. The pumps were calibrated before and after sampling.

Dust samples were also collected from each chamber at the nine time-points. At every elapsed time-point, one dust tray sample was collected from each chamber, which was covered with a Teflon lid and parafilm and stored at -20°C until further analysis. In addition, the wipe samples of the chamber interior surface (133 cm^2) were collected on the last day (day 30) by using a wipe cloth. A sterile wipe cloth ($5\text{ cm} \times 5\text{ cm}$) soaked with n-pentane by using the same procedure as that for standard dust was used. After sampling, the wipe samples were placed in glass jars, sealed with parafilm, and stored at -20°C until DEHP analysis.

To determine mold growth on the wallpaper samples at the nine time-points, we used ImageJ (version 1.52a) to calculate the mold growth area percentage on the wallpaper samples (Abramoff et al., 2004; Hoang et al., 2010). This mold growth percentage was also transformed to the American Society for Testing and Materials' (ASTM's) mold growth rating scale (Table 1).

2.4. DEHP analysis

The analytical procedure for DEHP in samples was modified from OSHA 104. Tenax and glass fiber filters were ultrasonically extracted with 5 mL of methanol/n-pentane (1:4). Next, 10 μL of internal standard (1000 ppm of benzyl benzoate; ChemService) was added. The extracts were pooled in a brown glass bottle, dried using N_2 gas, and reconstituted with 1 mL of methanol.

Each dust sample was fully mixed, and then 100 mg of dust was collected for DEHP extraction. This 100-mg sample was mixed with 10 μL of internal standard and then extracted with 2 mL of methanol and shaken ultrasonically for 30 min. After centrifugation at 2400 rpm for 10 min, the supernatant was transferred to a fresh glass tube. These steps were repeated twice; the supernatants were combined, dried using N_2 gas, and reconstituted with 1 mL of methanol. The same extraction procedure was applied to the wipe sample.

DEHP in the extracts was quantified using liquid chromatography-mass spectrophotometry (LCMS-8045; Shimadzu, Japan) with positive ion electrospray ionization (ESI+) and a Water XBridge C18 column ($3.5\ \mu\text{m}$, $2.1\text{ mm} \times 30\text{ mm}$). Mobile phase I was 0.1% aqueous ammonium acetate, and mobile phase II was methanol. The flow rate was 0.4 mL/min, and the sample injection volume was 1 μL .

The R^2 of the calibration curve ranging from 0.01 to 1.0 $\mu\text{g}/\text{mL}$ were 0.998. The limit of detection (LOD) was 0.176 ng/mL. In each batch analyzed, one blank, one quality check (QC) DEHP standard, and two spiked samples (0.1 g/mL DEHP) were also included. The recovery rates for the QC air, dust, and wipe samples were 89.15%–117.85%, 86.97%–116.57%, and 88.59%–116.38%, respectively; the recovery rate from the spiked air, dust, and wipe samples was 85.6%–100.2%, 70.66%–128.7, and 85.49%–122.01%, respectively.

Table 1. Fungal growth rating scale published by the American Society for Testing and Materials (ASTM).

Applied index	Observed fungal growth
0	No growth
1	Some growth detected, only with microscopy
2	Moderate growth detected, only with microscopy
3	Some growth detected visually
4	Visually detected coverage more than 10%
5	Visually detected coverage more than 50%
6	Visually detected coverage at 100%

2.5. Data analysis

The Mann–Whitney U test was used to verify the differences between each pair of experimental groups. A p of <0.05 was set as the level of statistical significance.

3. Results

3.1. Changes in temperature and RH during the experiment

The air temperature in the five chambers ranged from 26.86 to 29.63 $^{\circ}\text{C}$ (Figure 1a). The temperature CV of all five chambers at each sampling time-points as well as the temperature CV of the nine sampling time-points was $<5\%$. These results indicated that the temperature of the chamber during the experimental period was under control.

The RH of the five chambers during the experimental period was 94%–100% (Figure 1b). In particular, the RH in the chamber of moist WP, moist WP–AV, moist WP–CC, moist WP–PC, and moist WP–Mix was 97%–100%, 94%–97%, 94%–97%, 97%–100%, and 96%–99%, respectively. The RH CV of all five chambers at each sampling time-points was $<3\%$, and that the RH CV of the nine sampling time-points was $<2\%$. During the experiment, RH remained stable at $>94\%$, and the moisture content of the wallpaper samples in the five chambers at the nine sampling time-points was stable and high (51%–56%).

3.2. Mold growth

Mold growth was observed on all wallpaper samples with added mold but not on moist WP samples. As presented in Table 2, mold growth on wallpapers spiked with mold reached level 3, 4, and 5 growth on the ASTM scale on days 4, 8–15, and 15–25, respectively. The mold growth rate for the moist WP–CC and moist WP–Mix groups was the highest, whereas that for the moist WP–AV group was the lowest.

On the final day of the experiment (day 30), the area of wallpaper with mold growth was approximately 50% (47.29%–59.55%). The fastest mold growth was observed in the moist WP–Mix group ($58.22\% \pm 1.19\%$), followed by that in the moist WP–CC ($56.12\% \pm 2.08\%$), moist WP–AV ($51.51\% \pm 2.01\%$), and moist WP–PC ($48.72\% \pm 1.46\%$) groups.

3.3. DEHP in air

In the moist WP group, air DEHP concentrations decreased with time, and the highest concentration ($3.00 \pm 1.05\ \mu\text{g}/\text{m}^3$) was noted on day 2 (Figure 2a). However, the changes in DEHP emission from mold-added groups varied by mold species. DEHP emission from the moist WP–Mix samples stably increased from $1.71 \pm 0.29\ \mu\text{g}/\text{m}^3$ on day 2– $2.15 \pm 0.23\ \mu\text{g}/\text{m}^3$ on day 30 (Figure 2b). In the moist WP–AV and moist WP–CC groups, the highest air DEHP concentrations were detected on days 8 ($2.73 \pm 1.14\ \mu\text{g}/\text{m}^3$) and 10 ($2.76 \pm 0.66\ \mu\text{g}/\text{m}^3$), respectively. DEHP emitted from the moist WP–PC samples decreased with time, demonstrating a trend similar to that of the moist WP samples. In general, the moist WP samples released relatively more DEHP into air during the initial stages of the experiment. However, from day 8 onwards, relatively high DEHP concentrations in air were detected in all mold-added groups, except the moist WP–PC group.

The cumulative DEHP emission during the 30-day experimental period was calculated subsequently. The highest DEHP emission was noted from the moist WP–AV group ($4.61 \pm 0.55\ \mu\text{g}$), followed by that from the moist WP–PC group ($4.55 \pm 0.14\ \mu\text{g}$)—both were significantly higher than those emitted from moist WP ($3.87 \pm 0.27\ \mu\text{g}$; $p < 0.05$). Compared with the moist WP samples, the moist WP–Mix samples demonstrated higher cumulative DEHP emission ($3.94 \pm 0.85\ \mu\text{g}$), without statistical significance. In general, the cumulative DEHP emission from moist WP samples with added mold was higher than that from the moist WP samples without added mold; however, the moist WP–CC samples emitted less DEHP than did the moist WP samples (3.74 ± 0.73 vs. $3.87 \pm 0.27\ \mu\text{g}$).

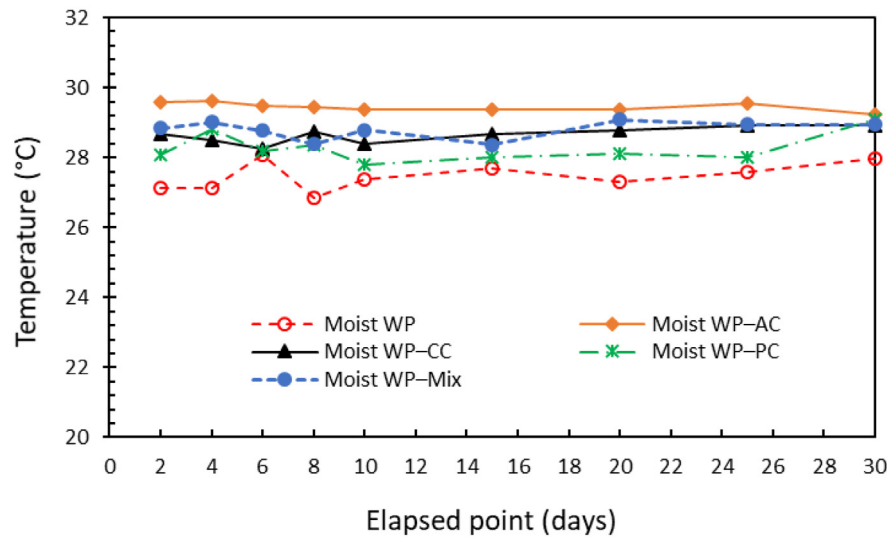
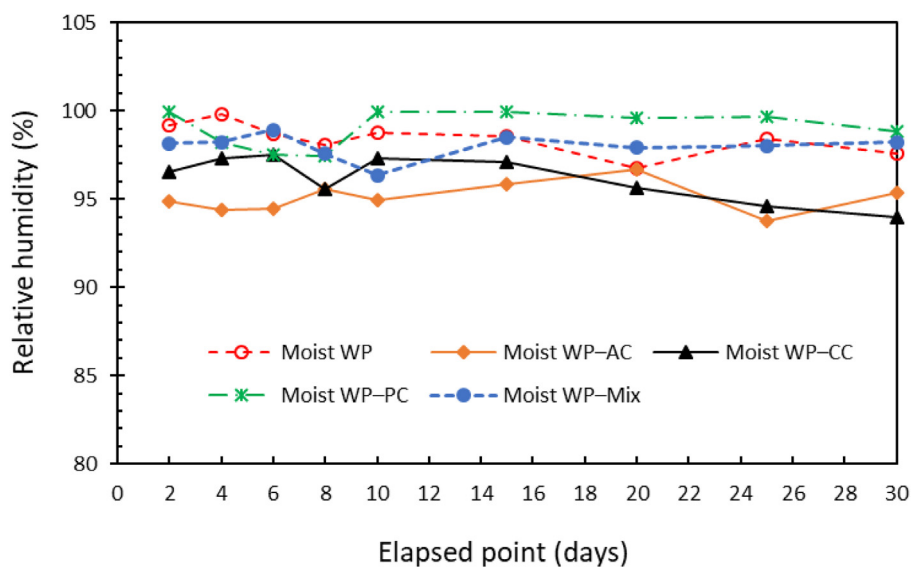
(a) Temperature**(b) Relative humidity**

Figure 1. Temperature and relative humidity at each time-point.

3.4. DEHP in dust

Figure 3 shows that the concentration of DEHP accumulated in dust increased with time. On day 2, DEHP concentration was the lowest in the moist WP group ($0.15 \pm 0.11 \mu\text{g/g}$) but the highest in the moist WP-AC ($0.83 \pm 0.13 \mu\text{g/g}$) and moist WP-PC ($0.87 \pm 0.71 \mu\text{g/g}$) groups. Between days 2 and 10, the moist WP-AC and moist WP-CC groups emitted higher DEHP concentrations into dust than did other groups ($0.53\text{--}1.39$ vs. $0.15\text{--}0.89 \mu\text{g/g}$). However, increases in dust DEHP concentrations in the moist WP-CC group became moderate on day 15. In the moist WP-PC and moist WP-Mix groups, DEHP concentrations increased after day 10. On day 15, dust DEHP concentrations became higher in the moist WP group ($1.39\text{--}4.63 \mu\text{g/g}$) than in the four groups spiked with mold ($0.90\text{--}4.01 \mu\text{g/g}$).

At the end of the experiment, the moist WP and moist WP-PC groups demonstrated the highest dust DEHP concentrations, whereas the moist WP-CC group demonstrated the lowest dust DEHP concentration (Figure 3). The cumulative dust DEHP concentrations were 11.37 ± 0.55 ,

12.00 ± 0.77 , 13.46 ± 0.71 , 13.63 ± 4.67 , and $13.93 \pm 0.49 \mu\text{g}$ in the moist WP-Mix, moist WP-CC, moist WP-PC, moist WP, and moist WP-AC groups, respectively.

3.5. DEHP in wipe samples

Among the wipe samples, DEHP concentration in the mold-added groups was significantly higher than in the moist WP group (Figure 4). The moist WP-PC ($3.06 \pm 0.65 \mu\text{g}$) and moist WP-Mix ($3.01 \pm 0.87 \mu\text{g}$) groups demonstrated the highest DEHP emission, followed by the moist WP-AC, moist WP-CC, and moist WP groups (2.08 ± 0.12 , 1.57 ± 0.33 , and $0.32 \pm 0.08 \mu\text{g}$, respectively).

3.6. Association of DEHP emission with mold growth

Because obvious mold growth was observed around day 10, the experimental period was divided into days 2–10 (D2–D10) and 15–30 (D15–D30) to evaluate the influence of mold growth on DEHP emission

Table 2. Fungal growth^a in the chamber system at nine elapsed times.

Elapsed point (day)	Moist WP-AV	Moist WP-CC	Moist WP-PC	Moist WP-Mix
2 nd	1	1	1	1
4 th	3	3	3	3
6 th	3	3	3	3
8 th	3	4	3	4
10 th	3	4	4	4
15 th	4	5	4	5
20 th	4	5	5	5
25 th	5	5	5	5
30 th	5	5	5	5

Moist WP: moist wallpaper.

AV: *Aspergillus versicolor*.

CC: *Cladosporium cladosporidias*.

PC: *Penicillium chrysogenum*.

Mix: the mixture of *A. versicolor*, *C. cladosporidias*, and *P. chrysogenum*.

^a By the fungal growth rating scale published by ASTM.

from wallpapers (Figure 5). Figure 5a illustrates that during D2–D10, the cumulative DEHP emitted into air in the moist WP group ($2.71 \pm 0.10 \mu\text{g}$) was higher than in the moist WP-CC and moist WP-AV groups ($1.94\text{--}2.49 \mu\text{g}$) without statistical significance ($p > 0.05$), but lower than in the moist WP-PC group ($2.94 \pm 0.07 \mu\text{g}$). The moist WP-PC group

exhibited a significantly higher cumulative DEHP emission onto dust during D2–D10 ($p < 0.05$). Contradictory results were obtained during D15–D30: the moist WP group contributed the lowest cumulative DEHP emission into air ($1.16 \pm 0.35 \mu\text{g}$; Figure 5b). Except for the moist WP-CC group, the differences between the moist WP groups with and without mold addition were significant ($p < 0.05$). During D15–D30, the highest DEHP concentrations were emitted in the moist WP-AV group ($2.12 \pm 0.21 \mu\text{g}$), followed by the moist WP-Mix group ($2.00 \pm 0.18 \mu\text{g}$).

For dust samples, contradictory results were obtained. Compared with the moist WP samples, those spiked with mold emitted more DEHP onto dust during D2–D10 ($5.35\text{--}9.64$ vs. $6.34 \mu\text{g}$; Figure 5c); however, the differences were nonsignificant ($p > 0.05$). Conversely, during D15–D30, the mold-added groups emitted less DEHP than did the moist WP group ($2.37\text{--}8.10$ vs. $7.29 \mu\text{g}$; Figure 5d); nevertheless, only the differences between the moist WP and moist WP-CC groups and between the moist WP and moist WP-Mix groups were significant ($p < 0.05$).

4. Discussion

In this study, DEHP emission from water-damaged wallpaper was associated with mold growth (Figure 5). Wallpaper is composed of paper and glue, both of which include nutrients suitable for mold growth (Bissett 1987). During growth, mold secretes extracellular enzymes to break down wallpaper and acquire carbon (Gaylarde et al., 2003; Viitanen et al., 2010). This leads to structural damage to wallpaper; during this decomposition, wallpaper's pore diameter and number increase, which enhances

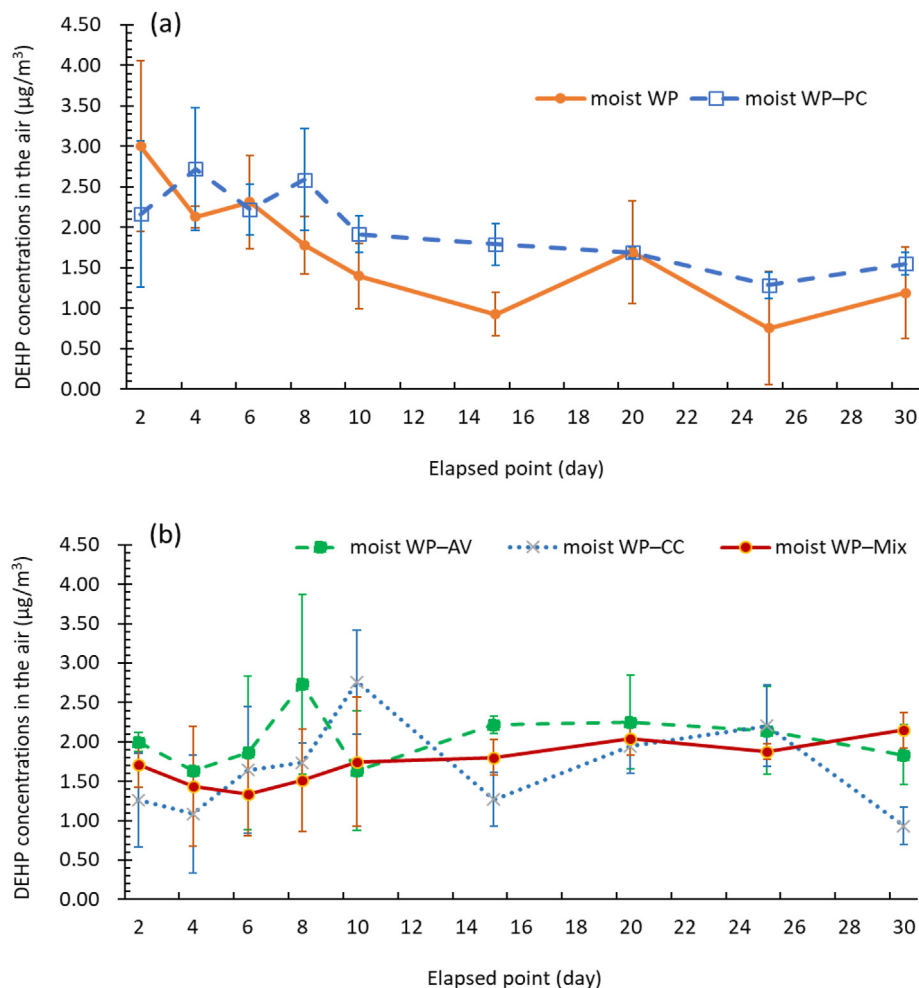


Figure 2. DEHP concentrations in air at nine time-points: (a) moist WP and moist WP-PC; (b) moist WP-AV, moist WP-CC, and moist WP-Mix. Moist WP: moist wallpaper; AV: *Aspergillus versicolor*, CC: *Cladosporium cladosporidias*, PC: *Penicillium chrysogenum*, Mix: the mixture of *A. versicolor*, *C. cladosporidias*, and *P. chrysogenum*.

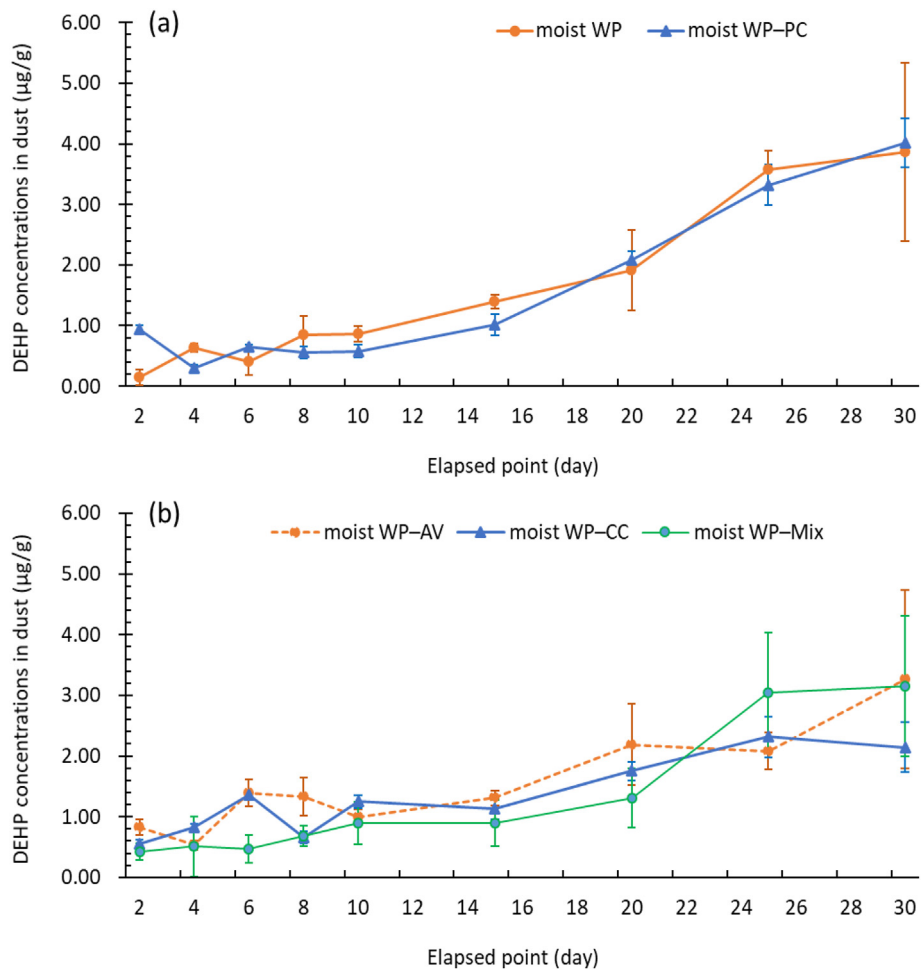


Figure 3. Cumulative DEHP concentrations in the dust at nine elapsed points: (a) moist WP and moist WP-PC; (b) moist WP-AV, moist WP-CC, and moist WP-Mix. Moist WP: moist wallpaper; AV: *Aspergillus versicolor*, CC: *Cladosporium cladosporidias*, PC: *Penicillium chrysogenum*, Mix: the mixture of *A. versicolor*, *C. cladosporidias*, and *P. chrysogenum*.

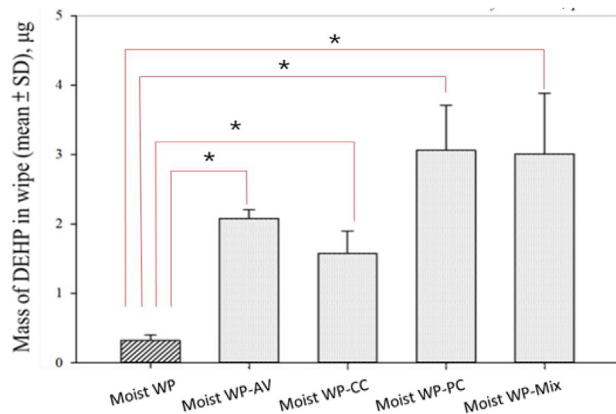


Figure 4. DEHP concentrations in wipe samples from the five chamber systems at the end of experiment. WP: Wallpaper; AV: *Aspergillus versicolor*, CC: *Cladosporium cladosporidias*, PC: *Penicillium chrysogenum*, Mix: the mixture of *A. versicolor*, *C. cladosporidias*, and *P. chrysogenum*.

DEHP emission (Viitanen et al., 2010). This mechanism underlies the moisture content-DEHP emission association. Water penetration and high moisture content in building materials destroy the structure of materials and accelerate their degradation, which favors DEHP movement from the interior of building materials to their surface (Miniotaite 2014).

Mold growth also lead to the production of various metabolites, such as microbial volatile organic compounds (MVOCs), hydrolytic enzymes, and acid, which cause material corrosion and breakdown (Gaylarde et al., 2003) as well as DEHP hydrolysis, thereby creating a DEHP concentration gradient. The DEHP concentration gradient between the interior and surface of materials increases DEHP migration to the material surface (Weschler 2004Weschler 2004), enhancing DEHP emission into the environment. For moist WP with mold growth, high moisture content is associated with a decrease in surface DEHP concentrations due to enhanced DEHP hydrolysis, which promotes DEHP concentration gradient formation. Given the effects of mold growth and high moisture content, wallpaper samples spiked with mold may demonstrate increased DEHP emission. Our current results demonstrated that during the experimental period, cumulative DEHP emission into the environment (dust, air, and wipe) was lower in the moist WP (without mold) group than in the groups with added mold (17.82 vs. 18.32–21.07 µg), except for the moist WP-CC group (17.31 µg). DEHP emission was the lowest in the moist WP-CC group potentially because of the change in RH in the chamber system. Over the 30 experimental days, RH in the chamber for moist WP-CC decreased from 97% on day 10 to 94% on day 30. The higher the RH is, the greater is the mold growth in the environment, such as an environment with dust (Dannemiller et al., 2017) or wallpaper (Nielsen et al., 2004). A RH decrease may be associated with *C. cladosporidias* growth; therefore, it may affect DEHP emission from the moist WP-CC. Figure 5b and 5d illustrate that during D15–D30 (visible mold growth), the moist WP-CC group demonstrated the lowest

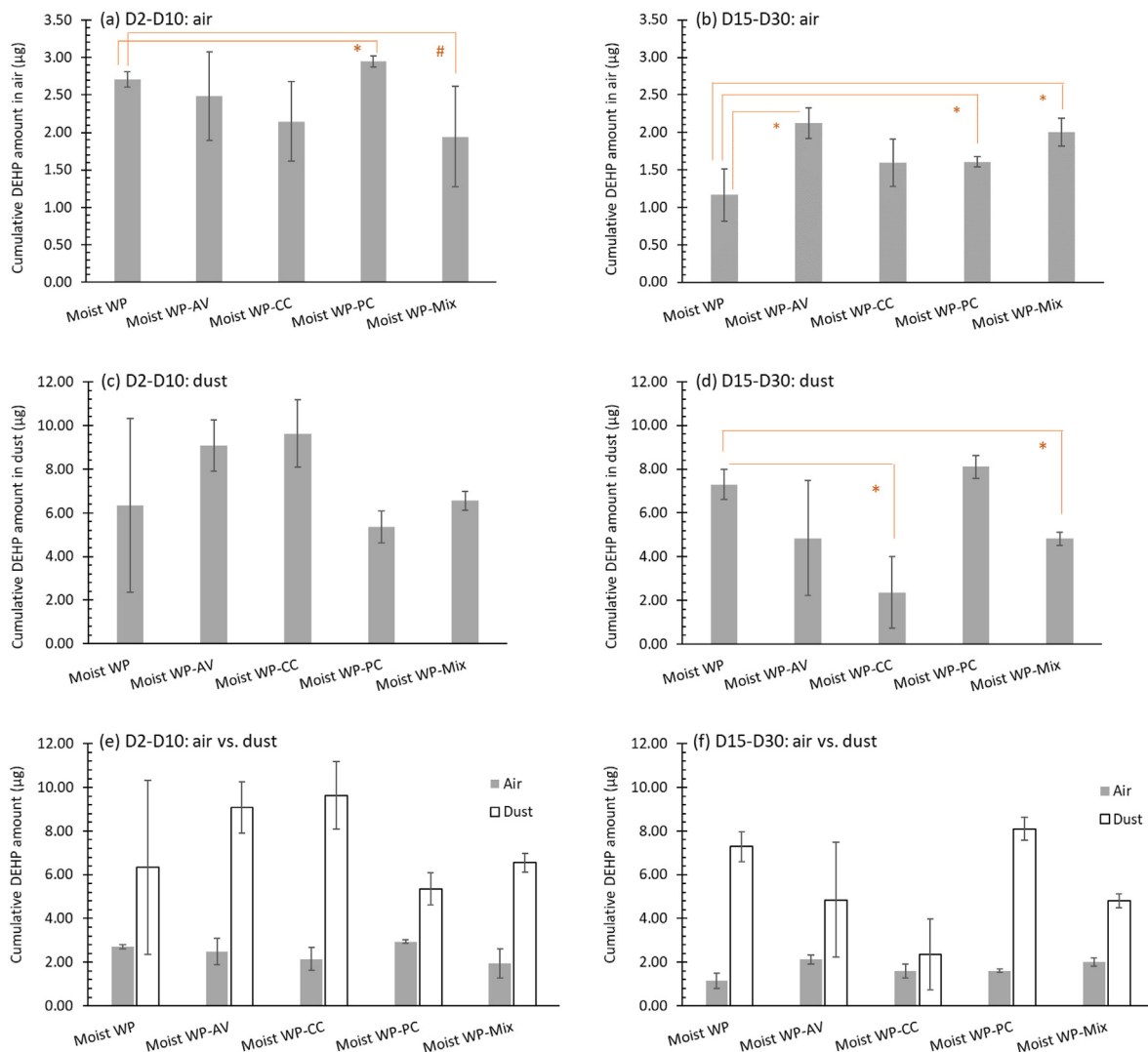


Figure 5. Cumulative DEHP amounts in air and dust among five chamber systems in the period without (D2–D10) and with (D15–D30) obvious mold growth: (a) D2–D10: air, (b) D15–D30: air, (c) D2–D10: dust (d) D15–D30: dust, (e) D2–D10: air vs. dust, and (f) D15–D30: air vs. dust. * $p < 0.05$ and # $p < 0.1$ by Mann-Whitney U test.

cumulative air and dust DEHP emission among all the mold-added groups. Figure 2b also demonstrates that the DEHP concentration in air seems to have a decreasing trend after day 10. Moreover, at the end of the experiment (day 30), DEHP concentrations in the wipe samples were the lowest in the moist WP–CC group (Figure 4).

We next compared the cumulative DEHP emissions between those during D2–D10 and D15–D30 to evaluate the influence of mold growth on DEHP emission into air and onto dust further. In the air samples, more cumulative DEHP emissions in the moist WP spiked with mold than in the moist WP group was only found during D15–D30 (Figures 5a and 5b). Studies have reported that airborne particles enhance DEHP emission into air because of the increased DEHP absorption area (Lee and Seo 2018) and convective mass-transfer coefficient (Benning et al., 2013). Mold spores are formed after mold growth (Sedlbauer, 2001); thus, more spores were formed and released into air during D15–D30 (with visible mold growth) than during D2–D10 (with no obvious mold growth). When DEHP was emitted from moist WP with added mold during D15–D30, airborne mold spore concentration was high, which potentially favored DEHP absorption into air; thus, cumulative air DEHP concentrations were higher in the mold-added moist WP samples than in the moist WP samples (Figure 5b). During D2–D10, mold growth was nonsignificant; therefore, the amount of mold spores in the air may have

been insufficient for DEHP absorption enhancement; thus, the DEHP emitted into air from moist WP spiked with mold was not higher than that from those from the moist-WP group (Figure 5a). Among the four mold-added groups, the cumulative DEHP emission into air was the highest from the moist WP–AV samples during D15–D30 (Figure 5b). This finding might be due to the spore size of *A. versicolor* (1.9–2.2 μm in diameter) being smaller than that of the other two mold species (*C. cladosporioides*: 2.3–2.5 μm ; *P. chrysogenum*: 2.6–3.0 μm ; Morris et al., 2000). The smaller a particle is, the larger is its adsorption-specific surface area; therefore, the smaller spore size of *A. versicolor* led to increased DEHP absorption and therefore increased the cumulative DEHP emission in air.

For dust samples, the moist WP–AV and moist WP–CC groups emitted more DEHP into dust than the moist WP samples during D2–D10 when the mold grew slowly (Figure 5c). However, during D15–D30, the cumulative DEHP emission from mold addition groups onto dust was relatively low (Figure 5d). Bope et al. (2019) reported that when $\text{RH} \geq 80\%$, DEHP in floor dust undergoes both abiotic and microbial degradation. For the mold-added groups, during D15–D30, mold was also noted in dust; therefore, degradation of DEHP in dust occurs through not only abiotic mechanisms but also biodegradation. This enhanced degradation in the mold-added groups led to decreased cumulative DEHP emission onto dust.

Here, we also observed that more DEHP was emitted onto dust (11.37–11.93 μg) than into air (3.74–4.61 μg) during our experimental period (Figure 5e and 5f); this result corroborates that of Clausen et al. (2004) and Shinohara and Uchino (2020). DEHP is a primary semivolatile compound; it tends to be adsorbed onto other interior surfaces and particles because of its low volatility (Shinohara and Uchino 2020). Therefore, even DEHP emitted into air might be deposited and transferred onto dust. Dust inhalation is the major nonfood route of DEHP exposure (occurring in 4.2–54.1% of different population groups), particularly in infants (54.1% formula-fed infants and 39.3% breastfed infants; Clark et al., 2003). Moreover, dust can be resuspended by wind or anthropogenic activities, increasing the possibility of DEHP inhalation from dust. Therefore, to control DEHP exposure, increasing cleaning frequency to minimize the amount of dust indoors is as crucial as reducing the use of PVC products and wallpaper and preventing dampness.

In the wipe samples, DEHP levels were higher in the mold-added groups than in the moist WP group (Figure 4). Because high RH possibly leads to the thick coverage of the surface by a water film and because DEHP has poor solubility in water and is nonpolar, the adsorption of DEHP onto the chamber surface is low (Chiou and Shoup 1985; Hippelein and McLachlan 2000). However, higher RH and the presence of water film on material surfaces provide suitable conditions for mold growth. When the spores together with DEHP land on the interior surface of study chambers, the spores may utilize water, reducing the interference of the water film in DEHP adsorption onto the chamber surface. However, the spores may begin growing and forming biofilms rich in organic matter and thus adsorbing DEHP. Fungal spores or biofilms with DEHP were also collected in the wipe samples. The main source of DEHP in the wipe samples may have been the spore particles and biofilms that had absorbed DEHP in the gas phase. This study is the first to evaluate the effects of mold growth on DEHP emission from moist WP. However, this study has two major limitations. First, even though numerous mold species may be detected in indoor environments, only three mold species were tested in the current study. Moreover, different mold species might lead to different rates of DEHP emission from wallpaper, as indicated by varying concentrations of DEHP emitted into air and onto dust in the wallpaper samples with different species of mold added (Figures 2, 3, and 4). Second, environmental conditions are more complex than those in our controlled chamber environment. Different microclimate and air pollution conditions as well as human activities and ventilation conditions can affect DEHP emission and distribution in air and dust and on material surfaces.

Our findings indicated that in contaminated environments, interactions occur between microbial growth and chemical emission. A moist indoor environment with mold growth can significantly contribute to DEHP emission. Thus, the role of moisture—a crucial factor inducing mold growth, which significantly contributes to DEHP emission and accumulation—warrants consideration. In addition, MVOCs, mycotoxins, and spores, produced and released into the environment during mold growth, are hazardous to human health (Kamijima et al., 2002; Tham et al., 2017; Wielogorska et al., 2019). Therefore, the multiple health effects of indoor environments with water damage, which fully represent human health risks in contaminated environments, require further investigation.

5. Conclusion

In this study, initially, the concentration of gaseous DEHP emitted from moist WP into air was higher than that from moist WP with mold; after at least 8 days, this trend reversed: moist WP with mold demonstrated emitted higher DEHP concentrations emitted into air. Regarding DEHP in dust, the concentrations were considerably increased 15 days after experiment initiation, particularly in the moist WP group. In general, mold enhanced DEHP emission from moist WP, particularly over D2–D10 for the dust samples and over D15–D30 for the air samples. More DEHP from moist WP was adsorbed onto dust than emitted into air.

Therefore, to control exposure to DEHP, minimizing the amount of dust indoors through frequent cleaning, reducing the use of PVC-containing products and wallpapers, as well as preventing dampness are essential. Moreover, from the public health perspective, removing or replacing moist WP after water damage, particularly that with obvious mold growth, is critical.

Declarations

Author contribution statement

Nai-Tzu Chen: Analyzed and interpreted the data; Wrote the paper. Ching-Hui Shih: Performed the experiments; Analyzed and interpreted the data.

Chien-Cheng Jung: Analyzed and interpreted the data. Nai-Yun Hsu: Conceived and designed the experiments; Analyzed and interpreted the data. Chung-Yu Chen; Ching-Chang Lee: Contributed reagents, materials, analysis tools or data.

Huey-Jen Su: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

Funding statement

Professor Huey-Jen Su was supported by Environmental Protection Administration, Executive Yuan, R.O.C. Taiwan [EPA-107-FA18-03-A176].

Data availability statement

Data will be made available on request.

Declaration of interest's statement

The authors declare no conflict of interest.

Additional information

Supplementary content related to this article has been published online at <https://doi.org/10.1016/j.heliyon.2022.e10404>.

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