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Characteristics and health risks of population exposure to phthalates via the use of face towels

Wenhui Zhang[1], *Na Zheng*[1,2,3,*], *Sujing Wang*[1], *Siyu Sun*[1], *Qirui An*[1], *Xiaoqian Li*[1], *Zimeng Li*[1], *Yining Ji*[1], *Yunyang Li*[3], *Jiamin Pan*[3]

[1] *College of New Energy and Environment, Jilin University, Changchun 130012, China*
[2] *Key Laboratory of Groundwater Resources and Environment, Ministry of Education, Jilin University, Changchun 130012, China*
[3] *Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences, Changchun 130102, China*

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ABSTRACT

The production of face towels is growing at an annual rate of about 4% in China, reaching 1.13 million tons by 2021. Phthalates (PAEs) are widely used in textiles, and face towels, as an important household textile, may expose people to PAEs via the skin, further leading to health risks. We collected new face towels and analyzed the distribution characterization of PAEs in them. The changes of PAEs were explored in a face towel use experiment and a simulated laundry experiment. Based on the use of face towels by 24 volunteers, we calculated the estimated daily intake (EDI) and comprehensively assessed the hazard quotient (HQ), hazard index (HI), and dermal cancer risk (DCR) of PAEs exposure in the population. PAEs were present in new face towels at total concentrations of <MDL–2388 ng/g, with a median of 173.2 ng/g, which was a lower contamination level compared with other textiles. PAE contents in used face towels were significantly higher than in new face towels. The concentrations of PAEs in coral velvet were significantly higher than those in cotton. Water washing removed some PAEs, while detergent washing increased the PAE content on face towels. Gender, weight, use time, and material were the main factors affecting EDI. The HQ and HI were less than 1, which proved PAEs had no significant non-carcinogenic health risks. Among the five target PAEs studied, DEHP was the only carcinogenic PAE and may cause potential health risks after long-term exposure. Therefore, we should pay more attention to DEHP.

Introduction

The textile industry has developed rapidly in recent years (Xue et al., 2017). The production of face towels in China is growing at an annual rate of about 4%, reaching 1.13 million tons by 2021 (CIR, 2021). In the textile industry, a series of complicated procedures will add softeners, plasticizers, color fixing agents, and other chemical additives, resulting in many harmful substances remaining in the textiles (Papaspyrides et al., 2009; Liu et al., 2017; Rovira and Domingo, 2019). Exposure to these chemical contaminants can cause various adverse impacts on the skin, such as the occurrence of *Pseudomonas* folliculitis (Teraki and Nakamura, 2015), contact dermatitis (Kawakami et al., 2014), skin microflora reduction (Walter et al., 2014), and other harmful health effects (Bocca et al., 2014; Bianco et al., 2015). Many

* Corresponding author.
E-mail: zhengnalzz@jlu.edu.cn (N. Zheng).

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studies have focused on the detection of inorganic and organic contaminants in textiles (Tuzen et al., 2008; Novick et al., 2013; Ionas et al., 2015; Avagyan et al., 2015; Luongo et al., 2014), the migration rates of contaminants in textiles during washing (Zhu and Kannan, 2020; Saini et al., 2016; Gong et al., 2016), the ecological risks of textile production, and possible health risks from textiles for children and workers (Reul et al., 2016; Tang et al., 2020; Ning et al., 2014, 2015; Checkoway et al., 2011). However, there is little information on the human health risks possibly associated with the use of face towels.

Phthalates (PAEs), a group of mass-produced plasticizers, are widely used in products such as textiles, personal care products (PCPs), and detergents (Guo and Kannan, 2013; Negev et al., 2018; Koppen et al., 2019). The addition of PAEs to textiles can improve their softness, flexibility, and durability (Net et al., 2015; Wang et al., 2018). Moreover, textiles can absorb organic substances in the environment (Morrison et al., 2016; Licina et al., 2019), which could also increase the PAE content of textiles. Nevertheless, PAEs are environmental endocrine disruptors that have estrogen-like effects and can bind to hormone receptors in the body, causing neurological disorders, endocrine disorders, and immunity reduction (Chang et al., 2021; Hlisnikova et al., 2021), leading to health problems such as reproductive and developmental impairment (Braun et al., 2013; Valvi et al., 2015; Zhang et al., 2019). Diethyl phthalate (DEP) has been found to strongly irritate the skin, conjunctiva, oral, and nasal mucosa (Mikula et al., 2005). Di(2-ethylhexyl) phthalate (DEHP) is significantly associated with reproductive problems, including early menopause and low pregnancy in women, preterm birth and low birth weight in infants, and decreased sperm quality in men (Wang and Qian, 2021; Chang et al., 2021). The U.S. Environmental Protection Agency (US EPA) has listed dimethyl phthalate (DMP), DEP, dibutyl phthalate (DBP), dioctyl phthalate (DOP), butyl benzyl phthalate (BBP), and DEHP as priority control pollutants (US EPA, 2014). In 2018, diisobutyl phthalate (DiBP) was added to restrictions in 28 EU countries (EU, 2018). Despite some countries have limited the use of PAEs, their annual use is still in the millions of tons (Net et al., 2015).

There have been several studies on PAEs in textiles, on topics such as face masks (Vimalkumar et al., 2022; Xie et al., 2022), sanitary napkins (Tang et al., 2019; Gao et al., 2020), preschool children's clothing (Tang et al., 2020), and jeans (Gong et al., 2016). However, the differences in contamination characteristics of face towels are often overlooked because their contact time with the skin is short. Nevertheless, the contact occurs more aggressively, as the wipe dries out wet skin, causing the potential release of pollutants (Rovira et al., 2017). Based on the detection frequencies of 16 PAEs, the list of priority contaminants (US EPA, 2014; EU, 2018), currently available toxicological data, and studies on PAEs in textiles (Li et al., 2019; Gong et al., 2016; Xie et al., 2022), we selected five PAEs as pollutants to monitor in this study, including DMP, DEP, DiBP, DBP, and DEHP. The objectives of this study were as follows: (1) to explore the distribution characteristics of PAEs in new face towels; (2) to investigate the contamination characteristics of PAEs in face towels used by 24 volunteers in a face towel use experiment; (3) to study the changes of PAEs during water washing and detergent washing in a simulated laundry experiment; (4) to evaluate the health risks of population exposure to PAEs via the use of face towels.

1. Materials and methods

1.1. Reagents

Conventional chemical reagents were n-hexane (HPLC grade) and acetone (AR grade) purchased from Aladdin (Shanghai, China). A standard mixture, containing five compounds DMP, DEP, DiBP, DBP, and DEHP at 1000 μg/mL in n-hexane, was obtained from Achemtek (purity > 98%). Appendix A Table S1 lists the physicochemical properties of the targeted chemicals. Ultrapure water used in the experiments was supplied by the reverse osmosis deionizer (BasicQ15, Hitech Inc., Shanghai, China).

1.2. Sample collection

Face towels can be made from cotton, Egyptian cotton, microfiber, solid cotton thread, coral velvet, or bamboo fiber. In September 2021, fifty face towels of each material were purchased from online retailers and local stores in Changchun, China. Based on the source (i.e., bought at a shop or online), the material, the price, and the density, thirty-one new face towels were randomly selected for contamination characterization. Details about the textiles are in Appendix A Table S2. The samples were enclosed in paper bags and stored at 4°C away from light.

1.3. Face towel use experiment

Researchers enrolled 24 volunteers in the study, all of whom signed informed consent forms. All volunteers were healthy and were asked to fill out a questionnaire on their use of face towels after the experiment. The questionnaire contains the use frequencies and approaches for the face towel and the frequency of use of personal care products (i.e., cleanser, skincare, sunscreen and whitening) by volunteers, and the results are shown in Appendix A Table S3. A chi-square test using SPSS vision 22.0 (IBM, Armonk, NY, USA) showed a significant difference in the frequency of use of PCPs between males and females ($p < 0.05$). By means of a pre-experiment to determine the PAE content of the new face towels, face towels made from different materials (i.e., cotton and coral velvet) were selected and given to the 24 volunteers. 12 volunteers (half of each gender) used face towels made of cotton for 4 weeks and the samples were recovered. They used the same face towels as before for another 6 weeks. The remaining 12 volunteers used face towels made of coral velvet in the same way as above. All the face towels were recovered after use. The volunteers could only use the issued face towels during the experiment.

1.4. Simulated laundry experiment

Researchers selected face towels made from different materials (i.e., cotton and coral velvet) with a relatively high PAE content and conducted seven rounds of simulated laundry experiments (Saini et al., 2016; Wang et al., 2019; Zhu and Kannan, 2020) to evaluate the migration rates of PAEs in face towels during washing. Before the simulated laundry experiment, 10×10 cm^2 square pieces of cloth (avoiding printed patterns) were randomly cut from selected face towel samples to determine the original PAE content. The details of the seven-round simulated laundry experiment are as follows.

(1) Water washing: A cotton face towel was cut in seven pieces (each piece about 10×10 cm^2), which were placed into seven 500-mL glass beakers, then 300 mL of water was added to each beaker, so the towels were just submerged. The beakers were shaken in an orbital shaker at a rate of 180 r/min for 15 min, then the towels were squeezed and dehydrated, and finally dried at room temperature. After the first round of washing, one of the seven pieces was taken as a sample for that wash. The remaining six pieces were washed for a second round in a similar fashion. When the second round of washing was completed, one of the remaining six pieces was taken as a sample for the second wash. Another piece of towel was removed after each subsequent wash as a sample for that wash. Each sample was separated into two portions and weighed to exactly 0.5000 g to produce parallel samples. A similar experiment was conducted with a coral velvet face towel.
(2) Detergent washing: Cotton and coral velvet face towels were selected as above, and washed in 300 mL of water and 3 mL of detergent (the main ingredients are listed in Appendix A). Samples were taken after each round as above.

1.5. *Determination of phthalates*

1.5.1. *Chemical analysis*

The towel samples were cut into small pieces and weighed to precisely 0.5000 g, then put into a 100-mL conical flask and 40 mL of n-hexane/acetone ($v/v = 1:1$) was added. The samples were extracted with ultrasound for 30 min, and the extraction solution was transferred to a conical flask. Then 30 mL of n-hexane/acetone ($v/v = 1:1$) was added, the ultrasonic extraction was repeated for 20 min, and all extraction solutions were combined. The extraction solutions were concentrated nearly to dryness by spinning in a water bath rotary concentrator (TurboVap 500, Biotage Inc., San Jose, CA, USA) at 38°C, then the concentrated solution was transferred to a graduated concentrate tube which had been washed with a little acetone. The concentrated solution was made to approximately 1 mL with n-hexane and passed through a 0.22-μm organic solvent-resistant microporous membrane. Finally, the solution was transferred to a brown 1.5-mL sample vial and analyzed by GC-MS (Li et al., 2015; Tang et al., 2020; Wang et al., 2021; Li et al., 2019).

1.5.2. *Instrumental analysis*

Gas chromatography (Agilent Technologies 7890 B) and mass spectrometry (Agilent Technologies 7000C) were used for the determination of PAEs. The separation was performed in selective ion monitoring mode using a fused silica capillary column (CD-5MS; 30 m × 0.25 mm I.D.; 0.25 mm film thickness). The sample (1 μL) was injected in a split stream. The ion source and inlet temperatures were 230°C and 280°C, respectively. The oven temperature was maintained at 40°C for 2.0 min, raised to 180°C at 10°C/min, then further raised to 350°C at 12.5°C/min. Mass spectrometry was performed with an electron bombardment ionization source (EI) as the ionization method and the system used a mass spectrometry detector (MSD).

1.6. *Quality control*

All containers were made of glass and rinsed with tap water and ultrapure water, then placed in an ultrasonic generator (KQ-500E, Kunshan Ultrasonic Inc., Kunshan, China) for 20 min. The containers were then washed three times with deionized water, baked in an oven at 60°C until no water droplets remained, cooled to room temperature, and finally washed twice with acetone. For each batch of twelve samples, three method blanks and three spiked blanks were processed throughout the sample preparation and analysis process. DMP, DEP, DiBP, DBP, and DEHP were detected with low levels in the method blanks, accounting for 0.05%–17.4% of the concentrations in real samples. In many previous studies on phthalate analyzation, low concentrations of PAEs were also found in blank samples (Tang et al., 2020; Li et al., 2019; Xie et al., 2022). Concentrations detected in the method blank were subtracted from the samples. All the concentrations of phthalates in samples were corrected with blank values. The GC-MS limits of detection were 0.2 ng/kg for DMP, DEP, and DiBP, and 1.4 and 3.7 ng/kg for DBP and DEHP, respectively. The standard spiked recovery experiment demonstrated that the recoveries of the five PAEs measured by the method were 83%–112%, which met the experimental requirements.

1.7. *Exposure risk assessment*

1.7.1. *Exposure time estimation*

Two hundred adult volunteers (> 21 years) including 100 males and 100 females were selected and they signed informed consent forms. The exposure time of the test subjects during face-washing was estimated by timing it with a stopwatch, beginning when the face towel was picked up with the hands, including the wiping of the face, and ending when the towel was put down. The geometric mean was selected as the contact time between the towel and the human body (Wilkinson and Lamb, 1999). The results are in Appendix A Table S4.

1.7.2. *Exposure dose evaluation*

Dermal exposures for adults (male and female) aged > 21 years were calculated using the equation based on the US EPA exposure assessment guidelines (US EPA, 2011).

$$\mathrm{EDI} = \frac{C_{\mathrm{towel}} \times d_{\mathrm{towel}} \times A_{\mathrm{skin}} \times F_{\mathrm{mig}} \times F_{\mathrm{contact}} \times F_{\mathrm{pen}} \times T_{\mathrm{contact}} \times n}{\mathrm{BW}} \times 10^{-3}$$

where EDI (ng/kg-bw/day) is the estimated daily intake via dermal absorption; C_{towel} (ng/g) is the concentration of phthalates in the face towel. d_{towel} (mg/cm^2) is the density of the face towel, the details are in Appendix A Table S2. A_{skin} (cm^2) is the contact area of the face towel and the human body, the estimated contact area for the face and hands for adult males and females was 1523 and 1270 cm^2, respectively (US EPA, 2011). F_{mig} (1/day) is the migration rate of phthalates to the skin, and the value is 0.005 (BfR, 2012). $F_{contact}$ (unitless) is the fraction of the skin contact area, and the value is 1 (BfR, 2012). F_{pen} (unitless) is the penetration rate of phthalates, and the value is 5% (BfR, 2012). $T_{contact}$ (day) is the contact time between face towels and skin, which is 13.7 sec for females and 9.3 sec for males (Appendix A Table S4). n (1/day) is the average number of events per day, and the value is 2. BW (kg) is body weightand 10^{-3} is the conversion factor.

1.7.3. Risk assessment

Based on RfD_d, the hazard quotient (HQ) was calculated as follows:

$$HQ = \frac{EDI}{RfD_d} \times 10^{-6} \quad (1)$$

$$RfD_d = RfD_{oral} \times ABS_{GI} \quad (2)$$

where RfD_d (mg/kg-bw/day) is the dermal absorption reference dose; RfD_{oral} is the oral reference dose, and the value is in Appendix A Table S5. ABS_{GI} is a gastrointestinal absorption factor, and the value is 1 (US EPA, 2018). 10^{-6} is the conversion factor.

HQ > 1 indicates a potential non-carcinogenic health risk.

The cumulative risk was estimated using the hazard index (HI), calculated with the following formula:

$$HI = HQ_{DMP} + HQ_{DEP} + HQ_{DiBP} + HQ_{DBP} + HQ_{DEHP} \quad (3)$$

HI > 1 indicates a chronic non-carcinogenic risk.

The dermal cancer risk of DEHP was calculated using the following formula (US EPA, 2004).

$$DCR = EDI \times SF \times 10^{-6} \quad (4)$$

where DCR is the dermal cancer risk; SF ($(mg/kg\text{-}bw/day)^{-1}$) is the cancer slope factor, and the value of DEHP is 0.014 (Wang et al., 2015). 10^{-6} is the conversion factor.

1.8. Statistical analysis

Statistical analysis was performed with SPSS vision 22.0 (IBM, Armonk, NY, USA). The Kolmogorov-Smirnov test was used to inspect the normality. The Mann-Whitney U test was used for comparisons between two groups when the data did not meet the normality. Statistical significance was set at $p < 0.05$.

2. Results and discussion

2.1. Relationship between the phthalate content and the characteristics of new face towels

The total concentration of PAEs in new face towels ranged from <MDL to 2388 ng/g, with a median of 173.2 ng/g (Table 1). The detection frequencies of DMP, DEP, DiBP, DBP, and DEHP were high (> 60%), suggesting that the face towels contained various phthalates, a finding similar to the results of others (Li et al., 2015; Xie et al., 2016; Li et al., 2019). DBP had the highest median concentration, followed by DiBP and DMP. However, DEHP (1278 ng/g) and DEP (489.4 ng/g) had the highest maximum concentrations. DEHP was the dominant contaminant in preschool children's clothing and infant cotton clothing (Tang et al., 2020; Li et al., 2019). Liu et al. (2020) found that the proportion of the 4 PAEs in new clothing was DBP (52.7%) > DiBP (24.2%) > DEHP (19.8%) > DMP (3.3%). For the new face towels, only some samples had a high DEHP content and the main contaminant was DBP. There were differences in the compositional distribution of PAEs for different textiles. The concentrations of PAEs in new face towels were lower than those measured in preschool children's clothing, some clothing from China, and infant cotton clothing (Tang et al., 2020; Chai et al., 2017; Li et al., 2019). Phthalates in jeans were the highest among the textiles studied, which may be related to exposure time and the local environment (Gong et al., 2016). Compared with other textiles (Appendix A Table S6), the new towels collected in the present study contained lower levels of phthalates.

DEHP, DiBP, and DBP are the most widely used chemicals in textiles (Rovira and Domingo, 2019; Li et al., 2019). In the global market, DEHP has been gradually replaced by diisononyl phthalate (DiNP) and diisodecyl phthalate (DiDP) (Tang et al., 2020). In this study, DEHP was detected in 80.65% of the samples, indicating that DEHP is still predominant in the manufacture of face towels. Moreover, it has been reported that DBP has been gradually replaced by DiBP (Dodson et al., 2012). However, the detection frequency and median concentration of DBP were higher than those of DiBP, indicating that both DBP and DiBP were used in face towels.

The mean concentration of phthalates varied with the source (i.e., bought at a shop or online), the material, the price, and the density (Table 1). The mean concentrations of DMP, DEP, DiBP, and DBP of the face towel samples bought online were lower than those of the shop-bought samples, but the mean concentration of DEHP was about 100-fold higher than that of the shop-bought samples. We found some samples bought online had a high DEHP content. Face towels bought online were sealed in plastic express packaging bags during transportation, and the bags also contained high concentrations of DEHP (Xu et al., 2020). Therefore, these face towels may absorb DEHP from the bags during transportation, which is an important source of PAEs.

Regarding the materials, the samples are divided into two categories: cotton and non-cotton (Wang et al., 2019). Non-cotton includes Egyptian cotton, microfiber, solid cotton thread, coral velvet, and bamboo fiber. Although the concentrations of PAEs in cotton were not significantly different from that in non-cotton ($p > 0.05$), the mean concentrations of PAEs were higher in cotton than in non-cotton. Previous studies have shown that PAEs are more easily adsorbed by cotton fabrics than by other fabrics (Saini et al., 2016, 2017).

No significant differences in the PAE content associated with the price were found ($p > 0.05$). The mean 5 PAEs content of the CNY 10–15 group had the highest with 782.5 ng/g, while the CNY 0–5 group was the lowest with 166.0 ng/g. The densities of face towel samples were normally distributed with a mean value of 36.28 mg/cm^2, so they were divided into 10–30, 30–40, and 40–60 mg/cm^2 groups. The mean 5 PAEs contents were respectively 172.2, 504.6, and 316.5 ng/g in 10–30, 30–40, and 40–60 mg/cm^2 groups, and there were no significant dif-

Table 1 – The concentration of phthalates in new face towels and the mean concentration of phthalates varied with the source, the material, the price, and the density (ng/g).

Characteristic	Categories	DMP	DEP	DiBP	DBP	DEHP	5 PAEs [a]
Total (n=31)	Minimum	<MDL [b]	<MDL	<MDL	<MDL	<MDL	
	Median	20.05	7.663	25.60	118.7	1.115	173.2
	Maximum	84.21	489.4	167.9	369.0	1278	2388
	GM [c]	30.18	11.33	60.47	93.90	2.898	198.8
	GSD [d]	24.41	90.01	53.09	105.8	315.6	588.9
Source	Shop (n=17)	30.00	41.65	64.19	152.8	2.005	290.7
	Online (n=14)	24.34	18.58	29.63	100.5	226.8	399.8
Material	Cotton (n=17)	30.54	43.26	63.56	150.2	122.7	410.2
	Non-cotton (n=14)	23.68	16.63	30.40	103.7	80.25	254.7
Price (CNY)	0–5 (n=11)	20.70	17.20	20.88	105.2	1.948	166.0
	5–10 (n=9)	27.96	73.27	64.54	137.4	3.052	306.2
	10–15 (n=6)	37.87	14.78	60.11	143.6	526.1	782.5
	≥15 (n=5)	28.86	6.17	66.96	149.7	0.61	252.3
Density (mg/cm^2)	10–30 (n=8)	20.84	22.84	19.64	106.6	2.302	172.2
	30–40 (n=10)	28.27	59.34	71.61	157.1	188.3	504.6
	40–60 (n=13)	30.87	14.78	48.68	121.6	100.6	316.5

[a] 5 PAEs = DMP + DEP + DiBP + DBP + DEHP.
[b] MDL = Method detection limit.
[c] GM = Geometric mean.
[d] GSD = Geometric standard deviation.

ferences according to the density ($p > 0.05$). But we found that the greater the density of a face towel, the higher the price.

2.2. *Contamination characteristics of phthalates in used face towels*

Purchased face towels made from cotton and coral velvet were distributed to volunteers for use, recovered and the concentration of PAEs was determined after use. Most of the volunteers used face towels twice a day, accounting for 83.33% (Appendix A Table S3). The use approach is to dry the face with a face towel after washing the face. The detection frequencies of DBP, DiBP, and DEHP were 100%, while DMP and DEP were respectively 85.11% and 95.74%. Total phthalate concentrations ranged from 0.25 to 58.85 µg/g, with a median of 3.929 µg/g. Phthalates with longer carbon chains, such as DBP, DiBP, and DEHP, were higher in used face towels, while DMP and DEP with shorter carbon chains were lower. This result is consistent with the findings of phthalate contamination in daily clothing (Liu et al., 2020). The total phthalate levels in used face towels were similar to those in infant cotton clothing (2.29–51.9 µg/g) (Li et al., 2019), preschool children's clothing (2.228–31.82 µg/g) (Tang et al., 2020), and clothing from China (2.063–48.32 µg/g) (Chai et al., 2017), and were 2–18-fold lower than in jeans (6.86–750.8 µg/g) (Gong et al., 2016), but much higher than in cotton fabrics (0.85–1.349 µg/g) (Saini et al., 2016). Appendix A Table S6 demonstrated a comparison of median concentrations of phthalates in previous studies.

Phthalates existed in both new and used face towels, and the contents of PAEs in used face towels were significantly higher ($p < 0.05$) (Fig. 1a). Liu et al. (2020) reported the difference of PAE content in the new and used clothing. Similar phenomena were found for contaminants such as bisphenol A (Wang et al., 2019). A major reason for the difference of PAE content in the new and used towels is the use of PCPs. Numerous people, especially the young, use a variety of PCPs, such as face cleansers, creams, and whitening products (Meng et al., 2021). PCPs contain a variety of phthalates (Li et al., 2022; Guo and Kannan, 2013; Huang et al., 2018), and they will accumulate on the used face towels. Furthermore, phthalates have been found in indoor air (Tri Manh et al., 2015, 2017; Bu et al., 2016). Face towels that are placed indoors after use can absorb phthalates from the surrounding environment (Morrison et al., 2015; Cao et al., 2016; Saini et al., 2017), particularly low molecular weight phthalates (ester side-chain, one to four carbons) such as DMP, DEP, DiBP, and DBP (Li et al., 2019). Similar phenomena were found for contaminants such as bisphenol A (Wang et al., 2019; Liao et al., 2012; Blanchard et al., 2014; Xue et al., 2016), polychlorinated biphenyls (Morrison et al., 2018), and flame retardants (Saini et al., 2016). Moreover, PAEs, especially DEHP and DiBP, were also present at different body locations (Gong et al., 2014), and were higher in the left and right palms compared to other body locations (Gong et al., 2016). PAEs present on the face and hands will accumulate on the face towels when wiping the face.

Phthalate contents in used face towels made from different materials were significantly different ($p < 0.05$) (Fig. 1b), indicating a strong correlation between phthalate content and fabric type (Liu et al., 2017; Tang et al., 2020; Xue et al., 2017). The concentrations of phthalates in coral velvet were significantly higher than those in cotton ($p < 0.05$). Coral velvet is composed of 85% polyester and 15% polyamide (Appendix A Table S2). There are about 300 million pounds of polyester and polyamide fibers recovered from plastic bottles every year (Kavilanz, 2016). Phthalates, such as DBP, BBP, and DEHP, have been detected in plastic bottles (Da Silva Costa et al., 2021). Clothing made of various synthetic fibers has higher levels of phthalate contamination (Tang et al., 2020). Furthermore, phthalates were possibly added to coral velvet during the manu-

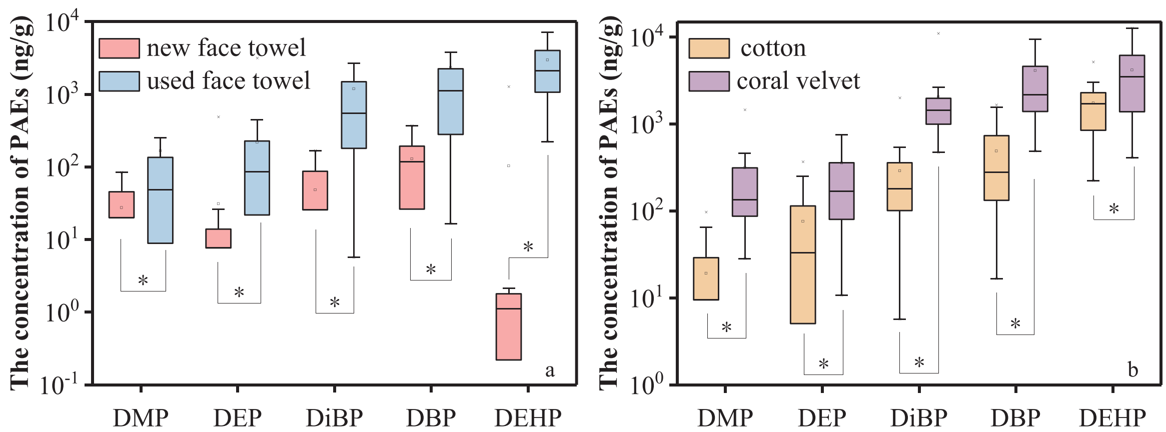

Fig. 1 – Box and whisker plots show the distribution of PAEs. Plot (a) represents the distribution of DMP, DEP, DiBP, DBP, and DEHP in new and used face towel samples. These were classified as new face towel samples (cotton [*n*=17], non-cotton [*n*=14]) and used face towel samples (cotton [*n*=24], non-cotton [*n*=24]). Plot (b) shows the distribution of PAEs in used face towels classified by material (i.e., cotton and coral velvet). The boxes represent the 25%–75% range, with the x signs representing the 1%–99% range and the error lines representing the 10%–90% range. The horizontal lines and squares inside the boxes mark the median and the mean, respectively. * $p < 0.05$.

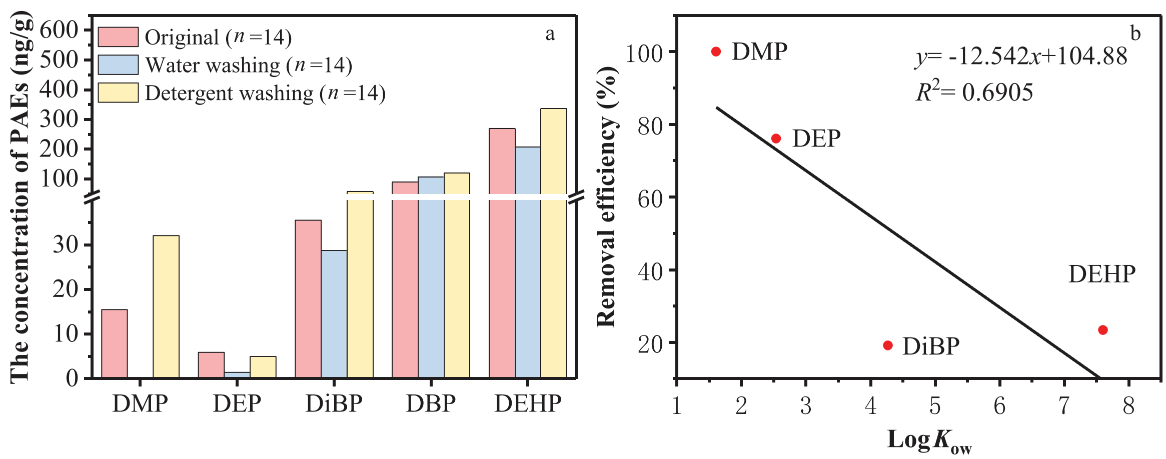

Fig. 2 – Plots of the simulated laundry process. Bar chart (a) represents the median concentration of PAEs in original, water washing, and detergent washing face towels. "Original" means the concentration of five target PAEs in the new face towel samples when they were not washed. Plot (b) represents the correlation between LogK_{ow} of phthalates and removal efficiency due to water washing. Values of LogK_{ow} are 1.61, 2.54, 4.27, and 7.60 for DMP, DEP, DiBP, and DEHP, respectively.

facturing to increase its softness (Net et al., 2015). The surface of coral velvet has a certain height of fluff, it has good water absorption, and phthalates are more likely to adhere to coral velvet compared to cotton during face wiping.

2.3. *Changes of phthalates in face towels during the simulated laundry process*

The changes of phthalates in face towels were studied in simulated laundry experiments (Fig. 2a). The concentrations of DMP, DEP, DiBP, and DEHP in face towels decreased after water washing. DEHP decreased most significantly, from 270.0 to 206.8 ng/g. Water washing removed some phthalates from the face towels, with respective removal efficiencies of 100.00%, 76.08%, 19.12%, and 23.38% for DMP, DEP, DiBP, and DEHP. As the octanol-water partition coefficient (K_{ow}) of a phthalate increased, the removal efficiency of water washing decreased (Fig. 2b), which is consistent with the fact that solubility in water is lower for a phthalate with a higher K_{ow} (Li et al., 2019; Gong et al., 2016). The LogK_{ow} values of PAEs are shown in Appendix A Table S1. The adjusted R^2 of a weighted linear regression between removal efficiency and LogK_{ow} is 0.69, suggesting a potential correlation between K_{ow} and the removal efficiencies of phthalates. Presumably, the correlation is related to the washing cycle during the laundering process. The concentrations of phthalates such as DiBP, BBP, and DEHP all decreased after water washing (Saini et al., 2016). PAEs were also found in the washing machine wastewater (Deshayes et al., 2015), suggesting that washing caused partial migration of PAEs from textiles to water, but standard laundry operations did not completely remove PAEs (Li et al., 2019; Gong et al., 2016; Negev et al., 2018). Although DBP and DiBP had the same LogK_{ow} value of 4.27, the removal rate of DiBP after water washing was higher than that of DBP. The possible reasons are as follows. On the one hand, some face towel samples adsorbed DBP from water during water washing. The adsorption rate of DBP from the water by the face towel was higher than the release rate of DBP from the face towel into the water. DBP was a typical PAE in water (Dominguez-Morueco et al., 2014; Zhang et al., 2011), and the concentration of DBP in water was significantly higher than DiBP (Hashizume et al., 2002; Shi et al., 2011). This was probably because DBP was the most worldwide used plasticizer all around the world (He et al., 2011). In order to simulate the real washing environment, the washing water was Changchun city tap water. Liu et al. (2014) investigated PAEs in source waters in China and found that DBP had high concentrations in groundwater in the northeastern region (Liao River Basin). On the other hand, the water solubility of DiBP is higher than that of DBP at a room temperature of 25°C (Appendix A Table S1). The percentage of accumulated chemical released to laundry water was higher for chemicals with high water solubility (Saini et al., 2016). Saini et al. (2016) investigated the distribution of the proportion of PAEs released to laundry water and left on fabric after washing and found that the proportion of DiBP released to laundry water was higher than that of DBP. Students of the university for development studies at Nyankpala Campus wash their face towels infrequently due to an irregular water supply on campus (Twumwaa et al., 2021), and some volunteers reported in the questionnaire that they washed their face towels very infrequently (Appendix A Table S3). This is undesirable because water washing can remove some phthalates. We suggest that users wash their face towels with water after use to reduce the content of PAEs in the face towels.

The median concentrations of DMP, DiBP, DBP, and DEHP in the face towels increased after detergent washing. Notably, potential contamination of face towels with phthalates could come from detergents, because many detergents are packaged in plastic and various phthalates are added during detergent manufacturing (Koppen et al., 2019). Furthermore, the migration of phthalates between the face towels and the surrounding environment during the drying process also affects the concentrations of phthalates in the towels. However, after washing infant cotton clothes and air-drying them for 4 hr, the contribution of PAEs by adsorption was low, accounting for 0.029%–3.54% (Li et al., 2019). The drying time in the simulated laundry experiment was less than 4 hr, therefore, the adsorption of phthalates by face towels from the surrounding air during drying can be ignored. The DEP concentration in the face towel samples decreased after detergent washing (Fig. 2a). A possible reason is that there was no high concentration of DEP in the detergent used in our experiment. During the washing process, the release rate of DEP from the face towel into water is higher than the adsorption rate of DEP from the solution by the face towel. There were significant differences for the DEP content of different detergents (Vinas et al., 2015). Cacho et al. (2015) determined DMP, DEP, DBP, and DEHP levels in four detergents, and low concentrations of DEP were detected in only one detergent.

2.4. *Dermal exposure analysis and cumulative risk assessment*

2.4.1. *Estimated daily intake*

A comparison of the exposure to PAEs among face towel users of different genders showed that the EDI values of DEP were significantly higher for females ($p < 0.05$). Although there were no significant differences for the other four PAEs, the mean EDI values of DMP, DiBP, DBP, and DEHP were higher for females than for males. A possible explanation for these results is that females use a greater variety of PCPs and at a higher frequency in daily life than males do (Meng et al., 2021; CDC, 2017). A survey of more than 2300 people in the U.S. showed that females use an average of twelve PCPs per day while males use six (Ficheux et al., 2015). The phenomenon was also evident from the questionnaire for 24 volunteers (Appendix A Table S3). The differences of DEHP and DiBP in male and female face towels were not related to the PAE content on their faces and hands. Because the levels of DEHP and DiBP were not significantly different between males and females at the same body position (Gong et al., 2014). In contrast, high concentrations of DEHP were found in the face towels of two male volunteers with a mean value of 3924 ng/g, who lived in the same room and rarely used PCPs. A possible reason is that the concentrations of DEHP in indoor air were higher for these two male volunteers, which might be associated with their preference to smoke indoors. We accounted for smoking in 24 volunteers (Appendix A Table S3), and only these two male volunteers regularly smoked in the room where the face towels were placed. Ma et al. (2022) reported that DEHP was the predominant PAE in tobacco with a mean concentration of 4.64 ± 6.21 mg/kg. Wang et al. (2020) investigated the influence of exposure to environmental tobacco smoke on phthalate exposure in pregnant women, and there were significant differences between pregnant women with or without exposure to environmental tobacco smoke. Therefore, there is a possible connection between the DEHP on the face towels of males and the smoke indoors.

Based on the exposure to PAEs among face towel users of different body weights, the lowest mean EDI was observed in the $>$ 65 kg group. For DEP, the EDI was significantly higher in the $<$ 50 kg group than in the 50–55 kg, 56–60 kg, and $>$ 65 kg groups ($p < 0.05$). This phenomenon is also consistent with a greater dilution effect on the absorption of PAEs in heavier individuals than in lighter ones (US EPA, 2011).

There was not a significant difference in EDI among volunteers who used face towels for different lengths of time ($p > 0.05$). However, the mean EDI of the population using face towels for 6 weeks was higher than for 4 weeks, suggesting that the longer the face towels are used, the easier it is for PAEs to accumulate on them, and the more harmful to humans they may become. PCPs accumulate on the face towels after use. Face towels that are placed indoors after use can absorb phthalates from the environment, and PAEs accumulate on the face towels as the use time increases. The longer the face towels are used, the more times they are wiped, and the more PAEs present on the face and hands will accumulate on the face towels. To mitigate the effects of PAEs in face towels on human health, users should replace them promptly.

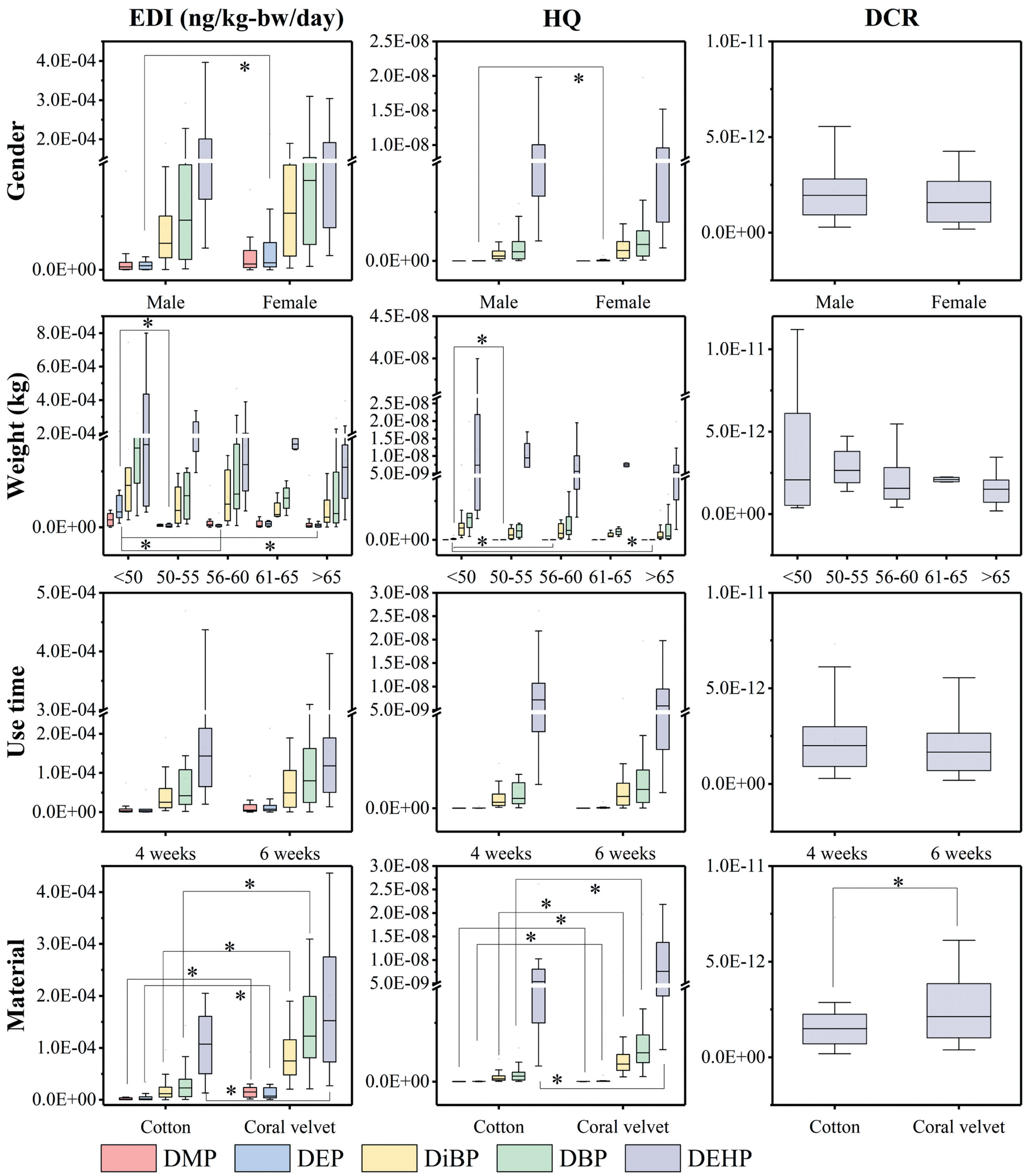

Fig. 3 – Box and whisker plots represent *EDI* and *HQ* of the population exposure to PAEs and *DCR* exposure to DEHP. The boxes represent the 25%–75% range, with the x signs representing the 1%–99% range and the error lines representing the 10%–90% range. The horizontal lines and squares inside the boxes mark the median and the mean, respectively. * $p < 0.05$.

Significant differences were found for different materials, the EDI values of PAEs were significantly higher for coral velvet than for cotton ($p < 0.05$). For DMP, DBP, DiBP, and DEP, the respective mean EDI values for coral velvet were 14-, 8-, 7-, and 4-fold higher than for cotton. The raw materials of coral velvet contain high concentrations of PAEs (Kavilanz, 2016; Da Silva Costa et al., 2021). Moreover, phthalates were possibly added to coral velvet during the manufacturing to increase its softness (Net et al., 2015). The surface of coral velvet has a certain height of fluff, it has good water absorption, and PAEs are more likely to adhere to coral velvet during face wiping.

2.4.2. Cumulative risk assessment

The hazard index method was used to estimate the simultaneous exposure to PAEs for face towel users. The regularities of HQ and HI for different genders, body weights, use time, and materials were the same as EDI. Among all the classifications, the mean HQ values of DMP, DEP, DiBP, DBP, and DEHP increased sequentially. The HQ values of DEP were significantly higher for females than for males ($p < 0.05$). A possible explanation is that females use a greater variety of PCPs and at a higher frequency in daily life than males do (Meng et al., 2021; CDC, 2017). DEP is the most common PAE in PCPs. Bao et al. (2015) measured levels of 11 PAEs in 198 PCPs collected from retail markets in Shanghai. DEP was the most frequently detected compound (29.8%) with a geometric mean concentration of daily exposure to DEP (0.018 µg/kg-bw/day). MEP is a metabolite of DEP in urine and higher MEP concentrations were associated with using PCPs (Fisher et al., 2019; Hsieh et al., 2019). Urinary concentrations of MEP showed a positive relationship with the number of PCPs used (Romero-Franco et al., 2011). Meanwhile, they were significantly higher in the < 50 kg group than in the 50–55 kg, 56–60 kg, and > 65 kg groups ($p < 0.05$). For each of the five phthalates, the HQ values of coral velvet were significantly higher than that of cotton ($p < 0.05$). The HQ values of DEHP were significantly higher than the other four compounds for different genders, body weights, use time, and materials ($p < 0.05$). Likewise, in studies of phthalate levels in indoor dust in China, Japan, and Korea, the most dominant phthalate was DEHP (Huang et al., 2021; Lee et al., 2021; Bamai et al., 2014), which indicates that in addition to the contamination of the face towels with PAEs during manufacturing, they could also be contaminated in other ways, such as by the absorption of dust during placement. The values of HQ were all less than 1, which means that there is no potential non-carcinogenic health risk of PAEs in face towels.

The median HI for PAEs was 7.33×10^{-9}, which is far below the safe value (HI = 1). The maximum HI was 6.74×10^{-8} for the face towel samples, indicating that the phthalates detected were all within the acceptable non-carcinogenic risk range (HI < 1). Similar results were found for other skin-contact products, such as infant cotton clothing (Li et al., 2019), diapers (Ishii et al., 2015), and sanitary napkins (Gao et al., 2020). The HI values for face towels are lower than for other products, mainly because the contact time between a face towel and the skin is very short, 13.7 sec for females and 9.3 sec for males, while the exposure time of PAEs in other products is measured in hours. However, face towels should not be ignored, because the frequency of replacing face towels is low, and as the use time increases, face towels are prone to accumulate PAEs from the surrounding environment and PCPs. The contact between a face towel and the skin occurs more aggressively, because the wipe dries out wet skin, further causing the potential release of PAEs.

2.4.3. Dermal cancer risk assessment

DEHP was present at the highest concentration among the phthalates studied and was the only phthalate with a carcinogenic risk. Previous studies have shown that a DCR value of 1×10^{-6} is considered an acceptable limit, which means that a DCR value greater than 1×10^{-6} indicates the possibility of a harmful impact (Maertens et al., 2008; US EPA, 2018). The mean and maximum DCR values for face towel users exposed to DEHP were 2.26×10^{-12} and 1.12×10^{-11}, respectively, which were much lower than 1×10^{-6}, indicating that the carcinogenic risk of a person exposed to PAEs via dermal contact through the use of a face towel is within acceptable limits. It further indicates that skin exposure to PAEs from a face towel has no significant effects on cancer incidence and mortality. Only some samples of other textiles, such as preschool children's clothing (Tang et al., 2020), face masks (Xie et al., 2022), and sanitary napkins (Tang et al., 2019), have DCR values near or above 1×10^{-6} and also have low dermal cancer risks. The carcinogenic risk values of infant cotton clothing are between 1.57×10^{-6} and 7.02×10^{-5}, and DEHP may cause potential adverse effects on infant health (Li et al., 2019). Although the dermal cancer risk evaluated in this study was minimal, just five PAEs were under consideration. The health risk of using face towels could be greater when exposed to various pollutants. In addition, this study only considered dermal exposure to PAEs in face towels; the carcinogenic risk would be increased if inhalation and oral routes of exposure were considered.

Fig. 3 shows that the mean DCR for female users (2.38×10^{-12}) is greater than that for males (2.13×10^{-12}), and the result is closely related to the fact that females prefer to use PCPs. A higher body weight is associated with a lower mean *DCR*, and a consequent lower cancer risk. The mean DCR when using face towels for 6 weeks was approximately equal to that when they were used for 4 weeks. Perhaps the users removed some DEHP that had accumulated on the face towels by water washing. Additionally, face towels placed in a room reached equilibrium with the PAEs of the indoor environment after a long while. Coral velvet towels presented a significantly higher cancer risk than cotton towels ($p < 0.05$).

3. Conclusion

Face towels play a significant role in dermal absorption exposure to PAEs. Phthalates existed in both new and used face towels, and the PAE content in used face towels was significantly higher. PCPs remain in face towels when towels are used for a long time. Most face towels are placed indoors, and PAEs in indoor air and dust can accumulate on them. Moreover, PAEs present on the face and hands will accumulate on the face towels when wiping the face. The concentrations of PAEs in coral velvet were significantly higher than those in cotton. The raw materials of coral velvet contain high content of PAEs. More phthalates were possibly added to coral velvet during the manufacturing to increase its softness. Coral velvet has good water absorption, and phthalates are more likely to adhere to coral velvet compared to cotton during face wiping. Water washing removed some PAEs, while detergent washing increased the PAEs on face towels. Potential contamination of face towels with PAEs could come from detergents. Because many detergents are packaged in plastic and various PAEs are added during the manufacturing of detergents. Gender, weight, use time, and material were the main factors affecting *EDI*. The HQ and HI values for DMP, DEP, DiBP, DBP, and

DEHP were far less than 1, demonstrating no significant non-carcinogenic health risks and chronic non-carcinogenic risks for the studied PAEs. The carcinogenic risk of DEHP was lower than 1×10^{-6}, with very low carcinogenicity after long-term exposure.

This paper focuses on the contamination characteristics of PAEs in face towels, which will help consumers raise their awareness of the safety of face towel use. To mitigate the effects of PAEs in face towels on human health, we suggest that users wash their face towels with water after use and replace them promptly. Our study only considered the dermal absorption route, and only the process of wiping the face after washing was considered for the exposure time estimation. Therefore, our calculation may be lower than the actual use. In the future, we plan to explore the accumulation of PAEs in face towels used for a longer time, compare the routes of exposure to PAEs through inhalation, oral ingestion, and dermal exposure, and assess the associated health risks.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this article.

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Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2022.10.016.

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