Chemosphere 219 (2019) 472–481

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Field evaluation of sequential hand wipes for flame retardant exposure in an electronics recycling facility

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HIGHLIGHTS

- Many factors affect wipe efficacy: material, number of sequential wipes, and lipophilicity.
- One hand wipe appears insufficient in quantifying total flame retardants on hands.
- Flame retardants likely remain on hands after 3 sequential wipes.
- Twill wipes might be better than gauze as a wipe material for flame retardants.

ARTICLE INFO

Article history:
Received 7 August 2018
Received in revised form
2 December 2018
Accepted 4 December 2018
Available online 5 December 2018

Handling Editor: Myrto Petreas

Keywords:
Flame retardants
Dermal wipes

ABSTRACT

Flame retardants have been associated with endocrine disorders, thyroid disruption, reproductive toxicity, and immunological interference. Through dismantling and recycling electronics and electric products, flame retardants can be released into the air and settle on work surfaces which may lead to dermal exposure. Hand wipe sampling is commonly used to evaluate dermal exposure. This study assesses the removal efficiency of wipes on the hands of recycling employees, and to compare the efficacy of two common surface wipe sampling materials. We used three sequential hand wipes and quantified the percentage of flame retardants that was removed by each hand wipe in the sequence. Two common wipe materials (gauze and twill) were used to compare the ability to remove flame retardants. The wipes were collected from 12 employees at a U.S. electronics recycling facility immediately at the end of their shift, prior to washing their hands. Results show that although the first wipe removed the highest median percent of the sum of the three wipes for most flame retardants, there was a wide range of the percentages of total individual flame retardants removed by both gauze (4%–98%) or twill hand wipe (1%–89%). Approximately half of the flame retardants a high percentage (>50%) removed by the second and third wipes. This suggests that a single wipe is not sufficient to characterize the extent of dermal contamination. The average of the total amount of flame retardants removed by twill wipes was greater than the average using gauze, but the difference was not statistically significant.

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1. Introduction

Flame retardants (e.g. polybrominated diphenyl ethers (PBDEs)) are commonly used in items to prevent or limit development of fire. These chemicals can be found in products such as carpets, upholstery, electronic goods, and many other consumer products. Although the toxicity of most flame retardants is not fully known, initial data indicates many of these compounds may pose a threat to human health. PBDEs are considered to be persistent organic pollutants as identified under the Stockholm Convention on Persistent Organic Pollutants [Stockholm Convention, 2015] and have a molecular structure similar to, and therefore may mimic,

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https://doi.org/10.1016/j.chemosphere.2018.12.027
0045-6535/Published by Elsevier Ltd.

thyroid hormones [McDonald 2002]. Some human epidemiologic studies have shown an association between exposure to PBDEs and changes in male reproductive hormones, semen quality, thyroid homeostasis, and hormone levels and fertility in women; cryptorchidism (undescended testicles); low birth weight and length; delayed motor skills; and decreased IQ [Dallaire et al., 2009; Czerska et al., 2013; Grant et al., 2013; Dishaw et al., 2014; Abdallah et al., 2015]. Although some PBDE formulations were phased out between 2004 and 2013, manufacturers have introduced replacements whose toxicity has not been well characterized [Allen et al., 2013]. These replacements include other brominated flame retardants i.e., 2-ethylhexyl 2, 3, 4, 5-tetrabromobenzoate (EH-TBB) and Bis(2-ethylhexyl) tetrabromophthalate (BEH-TEBP), and phosphorus flame retardants tris (1-chloro-2-propyl) phosphate (TCIPP) and triphenylphosphate (TPHP).

Electronics contains more than 1000 substances, many of which are hazardous [Wath et al., 2011], including heavy metals (lead, cadmium, mercury, beryllium, etc.), flame retardants, phthalates, and ozone depleting substances [NIOSH 2014]. Increasingly, end of life-cycle electronic products are being recycled as an alternative to landfilling. Serviceable components are removed and used in second generation products or materials such as plastics, precious metals, aluminum, copper, and other elements are harvested and re-sold as scrap. The potential exists for worker exposure to these hazardous substances during electronics recycling [Beaucham et al., 2017; Ceballos and Dong, 2016; Ceballos et al., 2016; Ceballos et al., 2015].

Hand wipes have been used to measure personal exposure to flame retardants in indoor environments. Levels of PBDE flame retardants on hands significantly correlated with serum PBDE levels [Watkins et al., 2011; Stapleton et al., 2012] and were a better predictor of serum PBDE levels than house dust [Stapleton et al., 2012]. In other studies, levels of some flame retardants on hand wipes significantly correlated with house dust concentrations [Hoffman et al., 2014; Stapleton et al., 2014]. In addition, hand wipe levels of tris (1,3-dichloro-2-propyl) phosphate (TDICPP), EH-TBB, and TPHP were significantly correlated with their urinary metabolites [Hoffman et al., 2014; Hoffman et al., 2015]. However, the majority of hand wipe studies have been conducted in homes or offices.

The industrial hygiene community has long used dermal wipe samples to evaluate exposure to workplace contaminants on the skin [Boeniger, 2007]. Surface and dermal sampling is especially appropriate for toxic substances that are persistent, solid or semi-volatile, and are able to bio-accumulate [Boeniger, 2007]. Flame retardants are considered semi-volatile and likely enter the body primarily through ingestion and dermal absorption [Abdallah et al., 2015]. Although assessments of wipe materials and number of wipes to assess dermal contamination has been done for some workplace skin contaminants [Ashley et al., 2009; Boeniger, 2007; Boeniger et al., 2008; Vostal et al., 1974], this type of assessment has not been done for flame retardants.

The National Institute for Occupational Safety and Health (NIOSH) performed a field study to evaluate a dermal wipe method to assess levels of flame retardant exposure by hand contact. This study had the following objectives: (1) to assess the need for repeat hand wipes to characterize the level of dermal contamination to flame retardants, and (2) to compare the performance of two commercially available wipe materials (gauze and twill) in measuring levels of flame retardants on the employees' hands. Most of the previous flame retardant dermal studies have been conducted in non-occupational settings with only one hand wipe. In addition, to the best of our knowledge, no studies have been conducted comparing two different wipe materials.

2. Materials and methods

2.1. Study design

To address objective 1, each participant wiped their hands with three sequential times at the end of their work shift, before they washed their hands at the end of the day. Each sample consisted of two wipes, one for the front of the hands and one for the back of the hands, following similar procedure used for flame retardant hand wipes by Carignan et al., (2013) and Ceballos et al., (2018).

To address objective 2 we evaluated two different wipe materials, gauze and twill. The wipe materials met the following criteria: (1) were commercially available, (2) had a low background concentration of flame retardants, and (3) were sufficiently rugged as to not disintegrate when collecting the sample. We selected gauze because it is typically used in flame retardant dermal wipe sampling studies. We selected the comparison wipe, twill, because they are typically used in cleanroom settings and have low background contamination. Wet wipes have been shown to be more efficacious than dry wipes in removing contamination from skin and other surfaces [Ashley et al., 2009]. We used isopropyl alcohol as our wetting agent because it is considered to be relatively non-toxic to the skin and did not contain the flame retardants for which we were sampling.

The research protocol used during this evaluation was approved by the NIOSH Human Subjects Review Board.

The flame retardants we analyzed are listed below. We used the abbreviation standard for flame retardants proposed by Bergman et al., [2012]. Please refer to Supplemental Table S1 for the definition and detection frequencies for these flame retardants.

2.2. Study population

This electronic recycling facility had 15 employees, including office and management personnel. The facility recycled a wide variety of electronic materials. Disassembly employees manually disassembled and sorted computer components, such as circuit boards, hard drives, copper wiring, and other parts that contained valuable materials. Employees also manually removed electronics from cardboard boxes and placed them onto a conveyor, which transported the electronics to a shredder (Fig. 1). A series of separators (including a magnetic separator and eddy current sorter) separated the scrap into individual components. We recruited 12 of

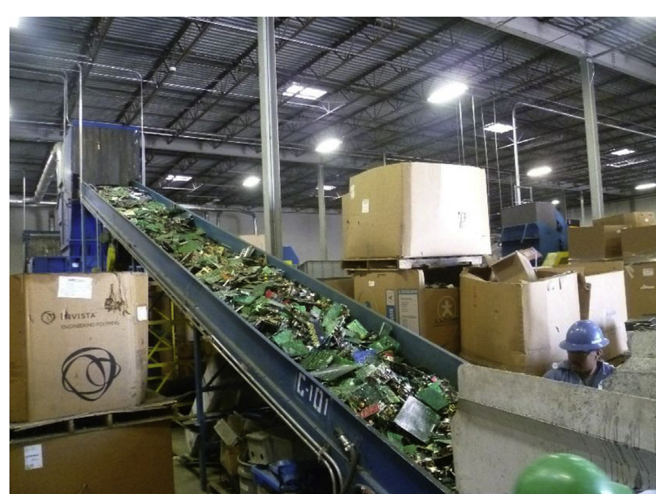


Fig. 1. Electronic shredding conveyor.

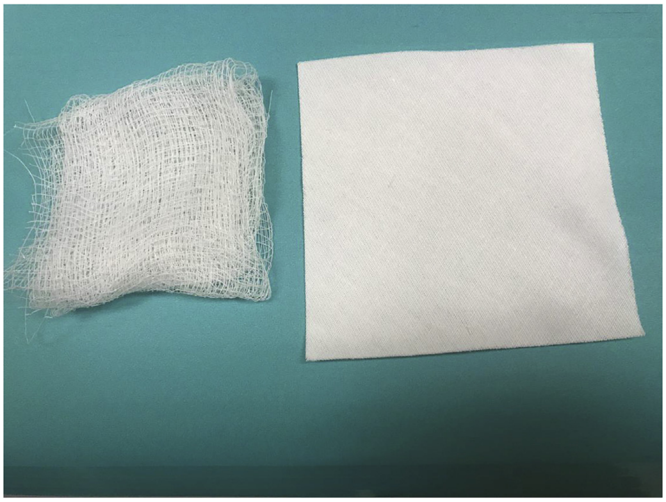


Fig. 2. Gauze pad (left) and twill wipe (right).

the employees to participate in our study, representing the two processing departments (disassembly and shredding). Six employees each used gauze or twill material for hand wipes. We grouped the employees so that the 6 employees using gauze had similar exposure to flame retardants as did the 6 employees that used twill (see Fig. 2).

2.3. Materials

Gauze pads (U.S.P. type VII, Dynarex, part number 3353) and twill wipes (MG Chemicals, part number 829–4x4) were presoaked with 99% laboratory grade isopropyl alcohol (Fisher Scientific, part number BP26324). Gauze pads were 3 × 3 inches, 100% cotton, loosely woven, and packaged in individual sterile packets. Twill wipes were 4 × 4 inches, 100% cotton, tightly woven, and packaged with 100 wipes per pack. Two of each type of wipe was placed into a labeled glass vial (Thermo Scientific, 120 mL Amber glass wide mouth jars, part number 340–0120) with 6 mL of isopropyl alcohol.

2.4. Methods

The hand wipe samples were collected immediately after the work shift, and before employees washed their hands at the end of the day. The NIOSH researcher donned a clean pair of nitrile gloves, opened the vial containing wipes, and directed the employee to remove one of the two wipes (either gauze or twill), wipe both of his or her palms from wrist to fingertips for at least 30 s, and then put the wipe back into the vial, following a standard practice for dermal wipes [ASTM 2013]. The wipe process was repeated with the second wipe for the back of both hands. After the employee placed the second wipe back into the vial, the researcher closed the lid, sealed it with parafilm, and placed the glass vial into a freezer at 0 °C. Each employee immediately performed the same process two more times. Each sample result consisted of analyzing both wipes (two wipes per sample). Sample results were reported as nanograms (ng)/sample. We also collected three field blank samples for each type of wipe. One blank contained only isopropyl alcohol and the other two each contained a pair of un-used gauze or twill wipes. In addition, one laboratory blank and one media blank per wipe type were retained in the laboratory for flame retardant analysis.

Hand wipes were stored frozen and shipped in a cooler with blue ice to the Virginia Institute of Marine Science (VIMS) laboratory for analysis. Once received at the laboratory all samples were stored (<4 °C) until analyzed. All hand wipes were analyzed for brominated and chlorinated organophosphate flame retardants by ultra-performance liquid chromatography atmospheric pressure photoionization tandem mass spectrometry using a method previously described by La Guardia and Hale (2015). All wipes were spiked with surrogate standards to determine the percent recovery. The surrogate standards were 500 ng of 2,3,4,4',5,6-hexabromodiphenyl ether (BDE-166) with a percent recovery mean of 90%, range 45–140%, 6000 ng of deuterated TDICPP, and with a percent recovery mean 104%, range 61–157% and 600 ng of deuterated TPHP with a percent recovery mean of 98%, range 48–191%. Media was spiked with the surrogate standards prior to extraction to determine extraction efficiency and to normalize the results for each type of flame retardant. All data are presented surrogate corrected for percent recovery. We found some of the flame retardants in the associated blank samples and list the detected flame retardants in Table 1. We compared the blank values to the sample results. It is likely that the media and containers that we used to ship samples to the lab likely contained TPHP and TCP. We collected and analyzed 2 lab, 2 media, and 2 field blanks per material. We also collected 2 isopropyl alcohol only blanks. We did not blank correct the results but provide the blanks with detected flame retardants for reference.

2.5. Data analysis

We used SAS® version 9.3 software for data analysis. For sample results that were reported as “not detected” we used the laboratory reporting limit divided by the square root of 2 [Hornung and Reed 1990]. The reporting limits for hand wipe samples was 63 ng per sample (ng/sample) for HBCDs, 156 ng/sample for TCEP, TCIPP, and TDICPP, and 10 ng/sample for the remainder of the flame retardants, where the sample consisted of the 2 wipes combined. We assessed the efficiency of the repeat hand wipes by calculating the percentage of each flame retardant that was removed by each wipe for each person. For each flame retardant sampled, we summed the amount of flame retardant removed in each of the three wipes, for each of the 12 participants. We then divided the amount removed on the first wipe sample, the second wipe sample, and the third wipe sample by the total amount removed by the wipes. We only included the data for participants that had all 3 sequential measures in this analysis. We calculated descriptive statistics and reported the medians and ranges for the flame retardants. We also compared the amount of flame retardants removed by each participant using the two wipe materials, gauze and twill. First, we added the flame retardants on the first wipe for each participant, and then compared the six gauze totals to the six twill totals. Next, we added the flame retardants on the first and second wipes combined for each participant, and then compared those six totals for gauze to the six totals for twill wipes. The totals were compared using Wilcoxon two-sample tests. If the p-value for a test was less than 0.05 the difference was considered statistically significant. We used Spearman correlation coefficients to examine the relationship between the quantity of flame retardants removed by the first wipe and the quantity of the flame retardants removed by all three wipes, for both types of material.

3. Results

Table 2 shows that the median and range of the percentage of the sum of flame retardants removed by the sequential hand wipes were similar for the gauze and twill hand wipe materials. We found that the first hand wipe removed the highest median percentage of the sum of flame retardants. Overall, the second and third

Table 1
Blank wipe sample results with detected flame retardants (in nanograms).

	Lab blank: gauze	Media blank gauze	Media blank twill	Field blank gauze 1	Field blank gauze 2	Alcohol only blank 1	Alcohol only blank 2
EH-TBB	7.5	ND	ND	ND	ND	ND	ND
BEH-TEBP	1.6	ND	ND	ND	ND'	ND	ND
TBB	ND	ND	1.6	ND	ND	ND	ND
TPHP	3.2	3.7	ND	4.4	5.3	1.4	6.4
TCP	7.7	3.2	ND	2.6	7.3	3.8	2.9

ND means not detected.

Table 2
Median and range of the percents of the sum of flame retardants removed by gauze or twill hand wipe samples.

	Gauze wipes (n = 86) Median of percents (range)	Twill wipes (n = 74) Median of percents (range)
Percent removed by 1st wipe	45 (4–98)	50 (1–89)
Percent removed by 2nd wipe	26 (1–55)	26 (5–93)
Percent removed by 3rd wipe[a]	23 (0–92)	23 (0–60)

[a] Due to laboratory analytical error we were unable to obtain results for 7 out of the 19 detected flame retardants on the third sequential wipes for 4 of the participants using gauze and 4 using twill. One of the twill participants was missing all of the measurements on the 3rd wipe.

sequential hand wipes both removed a similar median percentage of the sum of flame retardants, however when the second and third wipes are combined, the median of the percents for gauze was 55 and the median of the percents for twill was 50.

Table 3 shows the lower, median, and upper quartiles for the percent removed by the first wipe. Although the median percentile for gauze (45%) and twill (50%) were similar, the quartile range for the first gauze wipe was 23% versus the 18% for twill. Spearman correlation coefficients showed the quantity removed by the first wipe sample compared to the quantity removed by all three wipe samples. For gauze the correlation coefficient was 0.940 and for twill it was 0.980. Both were correlated with a p-value of <0.0001.

Fig. 3a shows the median of the percents of total flame retardants removed per gauze wipe for each of the employees that we sampled while Fig. 3b shows the median of the percents removed per twill wipe. We did not include the data for the fourth twill wipe participant because the third wipe sample was unable to be analyzed. For each employee, the first wipe for both types of wipe material, removed the highest median percent of the total flame retardants. However, the first twill wipes typically removed a greater median percent of total flame retardants than the first gauze wipes. We also found that the median percent removed by the third wipe was greater than that for the second wipe for two of the six employee participants using gauze wipes and one of the five employees using twill wipes. In addition, for five of the six participants using gauze wipes and two of the five participants using twill wipes the median percent of the total flame retardants removed by the combination of the second and third hand wipe was more than that removed by the first hand wipe.

We divided the hand wipe results for flame retardants by categories of use and years used in electronics [NIOSH 2017]. Table 4 shows this arrangement and the median level of flame retardants removed per wipe. Sources of flame retardants other than the electronic scrap, such as dust, conveyor belts, etc. were present in the processing areas and could have contributed to the overall exposure. In general, we found more flame retardants from electronic products that are either commonly used now and in the past (e.g., TCP, TPHP, TBBPA) and those commonly used in electronics until 2013 (e.g., DecaBDE, as indicated by BDE-209). BDE-209, which is the predominant congener for DecaBDE technical mixture [La Guardia et al., 2006], was frequently used in high impact polystyrene (HIPS) for television and computer monitor casings [Alaee et al., 2003]. Although its use was voluntarily phased-out of U.S. manufacturing and import by 2013, it is still found in many HIPS products produced prior to 2013 [EPA 2017]. TDICPP is not typically used in electronics, but we found relatively high median levels in the hand wipes. TDICPP is commonly used in polyurethane foam. Because polyurethane foam is used in so many types of products, its dust is found everywhere and can be pulled into electronic devices by their cooling fans.

The highest median level for the first gauze wipe was for TCP. TCP is found in cables, conveyor belts, plastics, and rubbers. The first twill wipe samples had a median of 2800 ng/sample (range 790–11,000) while the first gauze wipes had a median of 2600 ng/sample (range 810–7800).

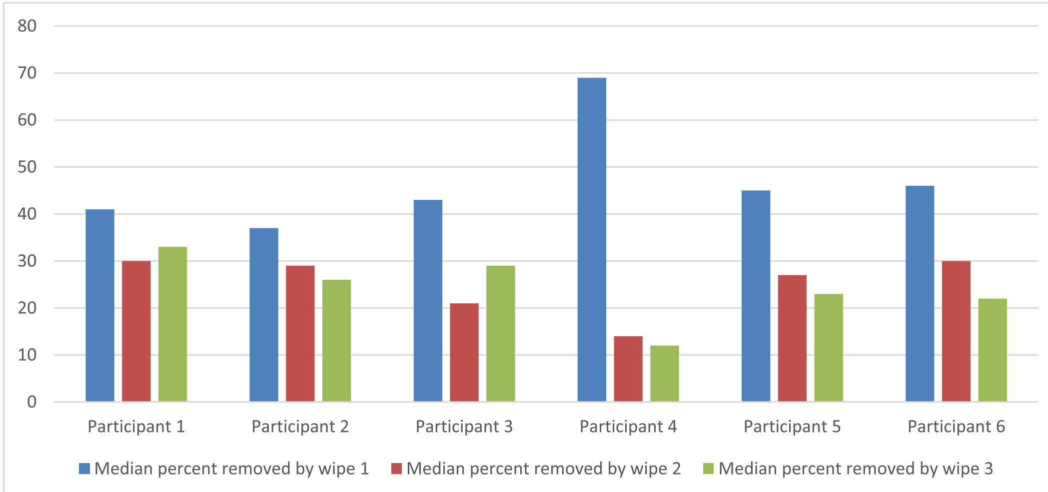
TBBPA had the highest median level for the first twill wipes and the second highest median level for the first gauze wipes. It is found in electronic circuit boards [Alaee et al., 2003], which were being disassembled and shredded during our evaluation. The median quantity removed in the first sample was higher on the twill wipes (4200 ng/sample (range 720–12,000) than in the gauze (2200 ng/sample (1000–6000). The subsequent sample showed almost triple the amount on the second twill wipe (median 2300 ng/sample, range 510–6200) than on the second gauze wipe (median 860 ng/sample, range 270–2900). However, the third wipe showed a median of 220 ng/sample (range 110–340) on the third twill wipe and a median of 510 ng/sample (range 81–930) on the third gauze sample.

BDE-209 also had some of the highest median levels. BDE-209 is the predominant congener for DecaBDE technical mixture [La Guardia et al., 2006]. DecaBDE technical mixture was frequently used in high impact polystyrene (HIPS) for television and computer monitor casings [Alaee et al., 2003]. Although its use was voluntarily phased-out of U.S. manufacturing and import by 2013, it is still found in many HIPS products produced prior to 2013 [EPA 2017]. The median of gauze handwipes for the first set of wipes was 2000 ng/sample (range 810–19,000) and 1500 ng/sample (range 310–17,000) for the second wipe sample. The third gauze handwipe had over three times the median level found on the first handwipe, 6700 ng/sample (range 330–21,000). The median for the first twill wipe sample was 4100 ng/sample (range 600–11,000), the second wipe sample was 1900 ng/sample (range 470–4600) and the third was 2200 ng/sample (range 470–4900).

Median levels of TPHP were approximately equal on the first twill and gauze wipes. TPHP, also commonly used in electronics including HIPS [Alaee et al., 2003], was found with a median of

Table 3
Lower (25%) median (50%) and upper (75%) quartile percents of the total flame retardants removed by wipe samples on the first wipe.

	Lower quartile % removed	Median % removed	Upper quartile % removed	Quartile range
Gauze (n = 86)	33	45	56	23
Twill (n = 74)	42	50	60	18



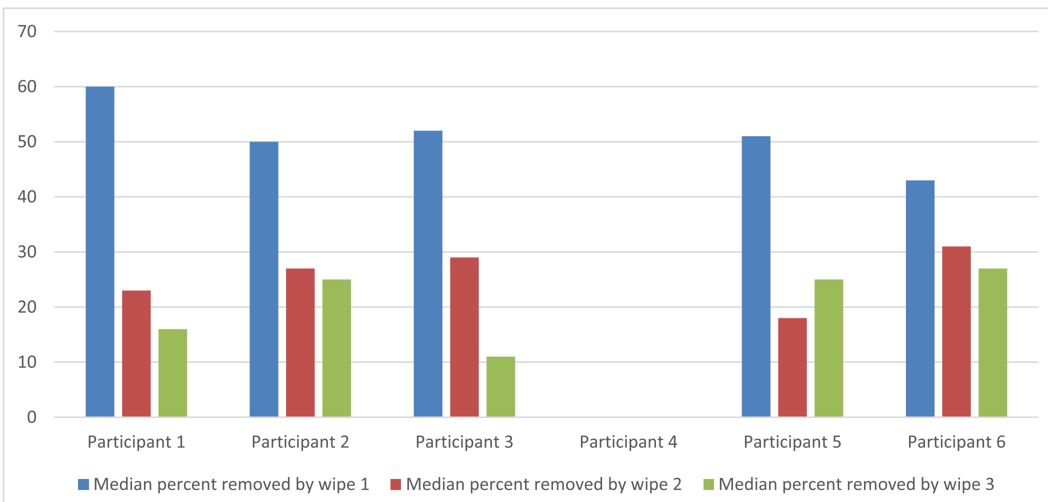


Fig. 3. a. Median of the percent of flame retardants removed by gauze wipes for each of the 6 participants. Participants 1 and 4 reflect data from all 19 of the flame retardants while participants 2, 3, 5, and 6 reflect data from 12 of the 19. Fig. 3b. Median of the percent of flame retardants removed by twill wipes for each of the 6 participants. Participants 1 and 2 reflect all 19 flame retardants while participants 3, 5, and 6 reflect data from 12 of the 19. We did not include the data for participant 4 because the third wipe sample was unable to be analyzed.

2800 ng/sample (range 990–12,000) in the first twill wipe sample and a median of 2100 ng/sample (range 580–5200) on the first gauze wipe.

The sum of all 19 flame retardants on the first wipe samples was 97,000 ng/sample for gauze and 170,000 for twill. We compared the sum of flame retardants on the first wipes for each of the six participants using gauze to the sum for the six using twill. For the gauze wipes the median of the six sums was 11,100 ng/sample (range 5900–35,000) and for twill was 26,100 ng/sample (range 4500–61,000). The difference in the amounts removed by the two materials on the first wipe was not statistically significant ($p = 0.32$, Wilcoxon 2-sample test).

The total quantity of flame retardants from the first and second wipe samples was 146,946 ng/sample on gauze ($n = 114$) and 295,634 ng/sample on twill ($n = 114$). We compared the amount of flame retardants removed on the first two gauze wipes for each of six participants to the amount removed on the first two twill wipes for six participants. The median amount removed for those using gauze was 15,300 ng/sample (range 9500–63,000) and the median amount removed for those using twill was 45,900 ng/sample (range 7600–120,000). Therefore, the twill wipes appeared to remove more flame retardants than the gauze wipes but the difference in the amounts removed was not statistically significant ($p = 0.40$, Wilcoxon 2-sample test).

The quantity removed in the first wipe significantly correlates with the quantity removed by all three wipes, for both gauze and twill ($p < 0.0001$, Spearman correlation coefficient). This indicates that the first wipe of either type of wipe is a good indicator of the total quantity of flame retardants per wipe.

For each flame retardant, Table 5 shows the median of the percent removed for each sequential wipe by type of wipe. For individual flame retardants, we found that the median percent removed by any of the three sequential wipes ranged from less than 1%–98%. It is also highly likely that additional flame retardants remained on the hands even after the third sequential wipe. For the gauze wipes, the median percent removed was highest on the first wipe sample 42% of the time, and on the sum of the second and third wipes 58% of the time. For twill wipes, the median percent removed was the highest on the first wipe sample 58% of the time, and on the sum of the second and third wipes 42% of the time.

The four flame retardants with the highest median levels removed on the first wipe were TCP, TBBPA, BDE-209, and TPHP.

Table 4
Median levels of each of the flame retardants removed per wipe sample in ng/sample, by type of wipe.

	Gauze			Twill		
	1st wipe sample n = 6 Median (Range)	2nd wipe sample n = 6 Median (Range)	3rd wipe sample n = 6 Median (Range)	1st wipe sample n = 6 Median (Range)	2nd wipe sample n = 6 Median (Range)	3rd wipe sample[a] n = 5 Median (Range)
OctaBDE (commonly used in electronics until 2004)						
BDE-183	52 (9.7–280)	35 (2.6–230)	34 (4.2–270)	87 (6.7–150)	35 (4.2–89)	43 (ND–79)
BDE-153	19 (5.2–39)	13 (2.9–28)	19 (ND–50)	28 (5.3–71)	15 (3.0–29)	27 (ND–42)
OctaBDE and DecaBDE technical mixtures (commonly used in electronics until 2013)						
BDE-209	2000 (810–19,000)	1500 (310–17,000)	6700 (330–21,000)	4100 (600–11,000)	1900 (470–4600)	2200 (470–4900)
BDE-206	63 (26–510)	41 (18–400)	ND (ND–1200)	160 (ND–310)	93 (15–170)	230 (ND–310)
Commonly used in electronics now and in past						
TPHP[b]	2100 (580–5200)	950 (260–1300)	590 (420–770)	2800 (990–12,000)	1700 (640–12,000)	350 (270–430)
TCP[b]	2600 (810–7800)	830 (200–3800)	1400 (110–2700)	2800 (790–11,000)	1600 (620–4900)	710 (540–880)
DBDPE	380 (110–1100)	190 (40–480)	210 (ND–2900)	320 (52–1900)	160 (42–470)	ND (ND–3500)
BTBPE	250 (120–690)	130 (12–680)	110 (ND–1200)	290 (25–960)	130 (16–280)	160 (ND–290)
TBBPA[b]	2200 (1000–6000)	860 (270–2900)	510 (81–930)	4200 (720–12,000)	2300 (510–6200)	220 (110–340)
Less commonly used in electronics now and in past						
EH-TBB	25 (20–110)	35 (18–150)	51 (31–320)	82 (44–480)	46 (25–300)	59 (23–280)
BEH-TEBP	39 (16–210)	46 (11–69)	89 (28–180)	58 (20–3800)	23 (9.7–2400)	130 (37–4700)
TCEP[b]	36 (ND–240)	ND (ND–130)	40 (ND–78)	84 (ND–1300)	69 (ND–280)	78 (71–84)
TCIPP[b]	220 (120–590)	150 (93–320)	170 (150–180)	540 (100–1700)	250 (150–920)	100 (75–120)
TBP[b]	15 (2.9–65)	3.2 (1.1–12)	ND	28 (6.2–78)	7.0 (4.2–18)	ND
Not generally used in electronics						
PentaBDE technical mixture						
BDE-99	62 (29–91)	38 (11–77)	25 (12–40)	125 (28–370)	58 (18–160)	25 (3.2–44)
BDE-47	38 (25–64)	26 (8.0–72)	23 (8.7–51)	65 (18–160)	39 (10–95)	35 (16–47)
BDE-100	8.1 (3.8–13)	6.1 (ND–14)	3.6 (2.3–8.0)	12 (3.7–46)	5.8 (3.0–19)	6.4 (3.6–9.3)
BDE-85	2.4 (0.96–4.6)	1.2 (ND–3.0)	ND	5.4 (ND–14)	2.3 (ND–8.6)	ND (ND–4.7)
TDCIPP[b]	510 (280–1700)	340 (150–530)	530 (380–680)	1100 (370–34,000)	560 (230–46,000)	41,000 (220–81,000)

ND = Not detected.
[a] The 3rd twill wipe for one of the participants was unable to be analyzed.
[b] These flame retardants had analytical errors in the 3rd wipe and therefore the n = 2 for the 3rd wipe for these flame retardants.

Table 5
Median percent of each of the flame retardants removed per wipe sample, by type of wipe.

	Gauze (n = 6)			Twill (n = 5)[a]		
	1st wipe sample Median (Range)	2nd wipe sample Median (Range)	3rd wipe sample Median (Range)	1st wipe sample Median (Range)	2nd wipe sample Median (Range)	3rd wipe sample Median (Range)
OctaBDE (commonly used in electronics until 2004)						
BDE-183	48 (30–83)	26 (6–47)	23 (11–36)	57 (41–61)	25 (17–36)	21 (6–28)
BDE-153	33 (25–77)	22 (18–30)	41 (5–54)	51 (41–59)	26 (16–33)	28 (8–33)
OctaBDE and DecaBDE technical mixtures (commonly used in electronics until 2013)						
BDE-209	41 (8–59)	18 (5–32)	33 (21–87)	49 (37–57)	26 (21–31)	25 (23–32)
BDE-206	54 (4–69)	35 (3–48)	1 (0–92)	32 (3–44)	19 (15–93)	43 (3–49)
Commonly used in electronics now and in past						
TPHP[b]	57 (43–70)	27 (18–35)	17 (12–21)	59 (55–62)	27 (26–27)	15 (11–18)
TCP[b]	68 (44–93)	19 (5–32)	13 (3–24)	54 (50–58)	32 (28–36)	14 (14–14)
DBDPE	42 (23–94)	15 (6–55)	22 (0–68)	55 (32–61)	39 (8–49)	1 (0–60)
BTBPE	52 (26–95)	27 (5–33)	22 (0–47)	60 (41–65)	26 (17–39)	18 (2–30)
TBBPA[b]	74 (61–88)	20 (9–30)	6 (3–9)	63 (52–73)	33 (26–40)	5 (1–8)
Less commonly used in electronics now and in past						
EH-TBB	27 (4–47)	26 (17–31)	45 (28–70)	46 (42–50)	26 (18–29)	26 (25–40)
BEH-TEBP	26 (11–55)	21 (18–26)	51 (27–68)	34 (33–45)	18 (12–22)	48 (33–55)
TCEP[b]	73 (48–98)	17 (1–32)	10 (1–20)	45 (1–89)	25 (5–44)	31 (6–56)
TCIPP[b]	40 (36–45)	39 (37–41)	21 (18–23)	54 (24–84)	29 (11–47)	17 (5–29)
TBP[b]	72 (67–77)	28 (23–33)	0 (0–0)	82 (77–86)	18 (14–23)	0 (0–0)
Not generally used in electronics						
PentaBDE technical mixture						
BDE-99	47 (38–63)	32 (13–42)	18 (15–34)	65 (48–71)	27 (25–38)	7 (4–25)
BDE-47	41 (31–58)	30 (14–47)	24 (17–45)	48 (45–60)	27 (24–40)	24 (15–25)
BDE-100	42 (32–59)	36 (8–47)	20 (13–39)	49 (42–66)	26 (18–38)	22 (9–40)
BDE-85	57 (40–64)	29 (18–44)	15 (8–30)	63 (46–66)	23 (18–50)	13 (4–21)
TDCIPP[b]	41 (41–42)	21 (16–26)	38 (33–42)	36 (21–50)	31 (28–33)	34 (17–50)

[a] The 3rd twill wipe for one of the participants was unable to be analyzed.
[b] These flame retardants had analytical errors in the 3rd wipe and therefore the n = 2 for these flame retardants.

The percentage of BDE-209 removed in the first wipe sample (relative to the sum total of all three wipe samples) was low. Only a median percent of 41% (range 8–59) was removed with gauze and a median percent of 49% (range 37–57) was removed with twill. The median of the percents removed for the first TBBPA wipe sample for gauze and twill was similar, with a median percent of 74% (range 61–88) for gauze and a median percent of 63% (range 52–73) for twill. The median percentage of TPHP removed by the first wipe

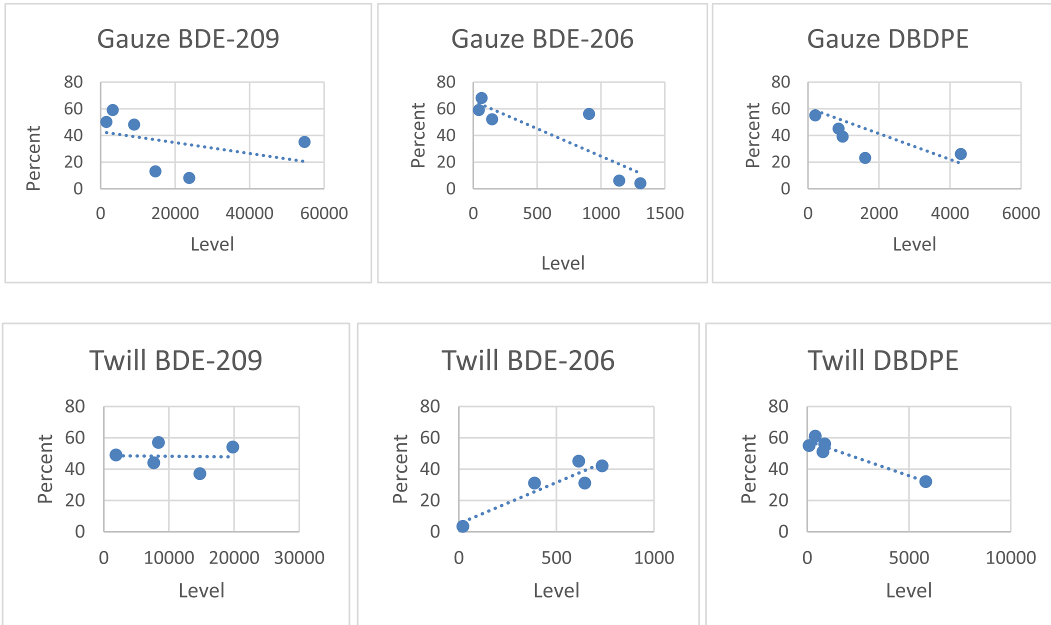


Fig. 4. a. Select scatterplots for Gauze wipe samples comparing quantity removed with the percent removed in the first wipe. Fig. 4b. Select scatterplots for Twill wipe samples comparing quantity removed with the percent removed in the first wipe.

sample was also similar for gauze 57% (range 43–70) vs twill 59% (range 55–62). The median percent of TCP removed in the first wipe sample had a greater range for gauze (median 68% [range 44–93]) but not for twill (median 54% [range 50–58]).

Fig. 4a and b shows selected scatterplots with trend lines showing the association between the percent removed in the first wipe and the sum of each specific flame retardant on all three wipes (in ng/sample). We examined these data to evaluate whether the total amount of flame retardant on the hands had any relationship with the percent removed by the first wipe. We found mixed results. We could not evaluate TPHP, TCP, TBBPA, TCEP, TCIPP, TBP, or TDCIPP because of insufficient data. Two of the flame retardants (BDE-85 and BDE-100) did not have a sufficient range of levels to adequate evaluate this concept. Most of the gauze wipe samples (BDE-47, BDE-99, BDE-183, TBB, and BTBPE) showed minimal evidence that the higher the level of the flame retardant the lower the percent removed in the first wipe. TBPH and BDE-153 showed no relationship and BDE-206 and DBDPE showed an apparent negative relationship between level and percent removed in the first wipe. BDE-209 showed a slightly negative relationship between percent removed and level in the first wipe. Twill wipes on the other hand did not show any relationship for levels of any of the flame retardants with percent removed in the first wipe. The apparent positive relationship in the BDE-206 twill wipes and the apparent negative relationship in the DBDPE twill wipes are due to one wipe sample.

We also arranged the flame retardants by their octonol/water partition coefficient (Kow) values as described by Schreder and La Guardia (2014). The K_{ow} value is defined as the ratio of a chemical's concentration in the octanol phase to its concentration in the aqueous phase of a two-phase octanol/water system. Fig. 5a plots the median of the percents of the specific flame retardant removed in our three sequential gauze hand wipes by ascending log K_{ow} values. As previously noted, the first wipe typically removed the majority (>50%) of the flame retardants. However, for flame retardants with log K_{ow} >7.5, flame retardant removal levels by the third wipe exceeded the first wipe for 33% of the flame retardants (i.e., BDE-153, EH-TBB, and BEH-TEBP) and 55% of the flame retardants removed by the third wipe exceeded those detected in the second wipe (i.e., BDE-153, BDE-209, EH-TBB, BEH-TEBP, and DBDPE). TDCIPP (log K_{ow} of 3.27) followed a similar pattern to DBDPE and had the most flame retardant removed on the first wipe, followed by the third wipe, and lastly by the second wipe. Fig. 5b plots the median percents of the flame retardants removed in the three sequential twill wipes. Twill wipes showed that BDE-206 (log K_{ow} 11.2) and BEH-TEBP (log K_{ow} 11.95) had the largest amount of flame retardant detected in the third wipe, followed by the first wipe and second wipe, whereas all other flame retardants had the largest amount detected in the first wipe, followed by the third wipe.

4. Discussion

One objective of our pilot study was to assess the need for repeat hand wipes to best characterize the level of skin contamination. Very little is known about the ability of a single wipe to characterize skin contamination of flame retardants. However, research on dermal exposure to lead have suggested that a single wipe may not be sufficient to characterize skin contamination [Boeinger, 2007; Vostal et al., 1974]. Boeinger, [2007] dosed hands with lead oxide, and then used three different wiping materials to determine total recovery. Overall, the best recovery found in the first wipe of the series was only 62%. When three consecutive hand wipe samples were analyzed, only 75% of the total dose was recovered. Boeniger found at the high loading level, indicated that the higher the dose of lead, the less efficient the first wipe. The Boeinger et al. study was similar to a previous hand wiping study by Vostal et al., [1974] who found approximately 69% of lead was removed in the first wipe and 31% in the second. Our hand wipe results have similarly shown insufficient recovery from a single wipe.

To our knowledge, there has never been an absolute recovery study, where a known quantity of flame retardant is applied to the skin to test the percent recovered with wipes. We performed a relative recovery study, which is based on the amount obtained from a subsequent consecutive wipes of the same surface area [ASTM 2003]. When a sampling method recovers a predominant

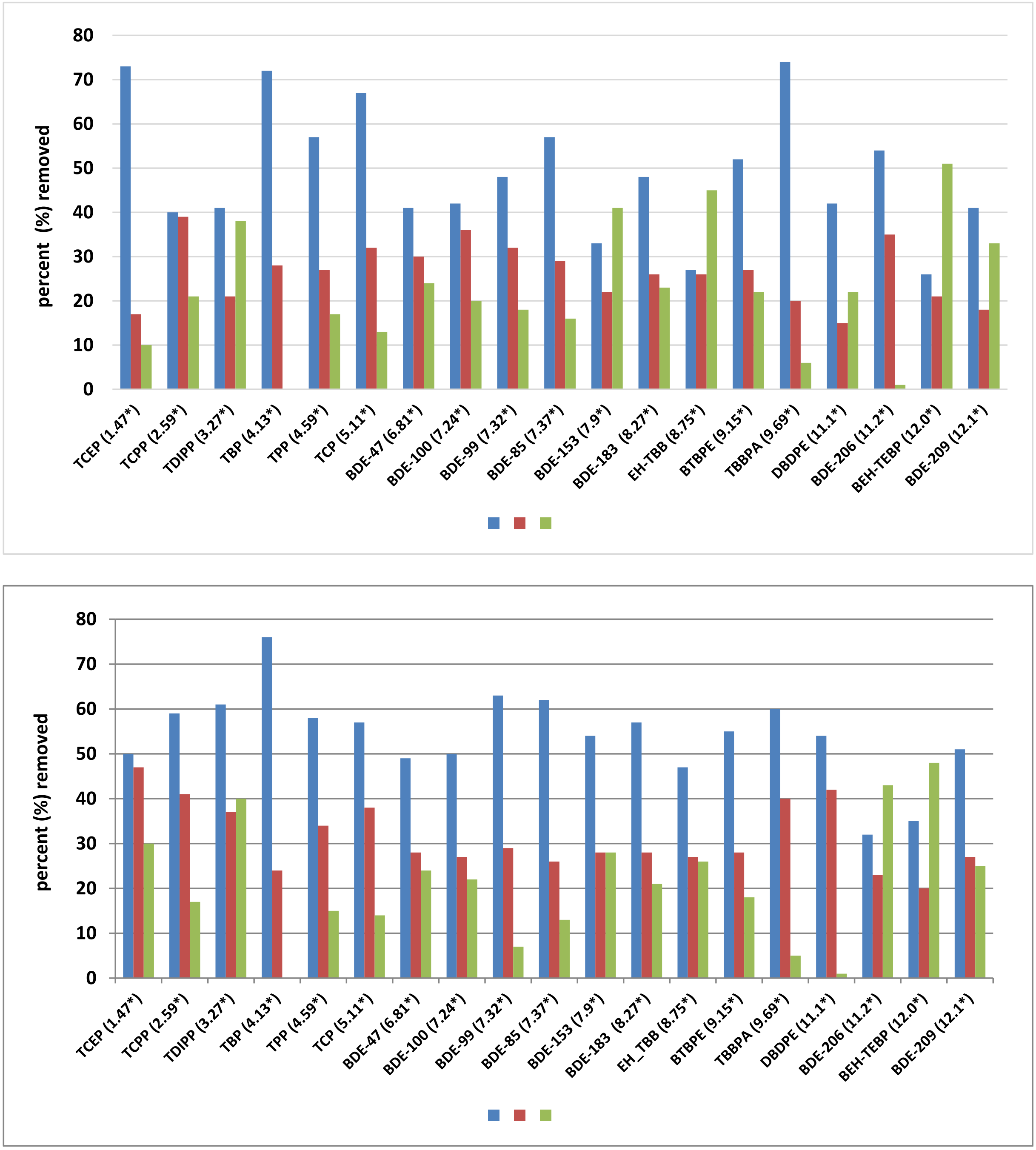


Fig. 5. a. Median percent flame retardant removed by sequential gauze wipes, flame retardants listed by increasing log octanol/water partition coefficient (log K_{ow}) values, range 1.47–12.1 log K_{ow}. Fig. 5b. Median percent flame removal removed by sequential twill wipes, flame retardants listed by increasing log octanol/water partition coefficient (log K_{ow}) values, range 1.47–12.1 log K_{ow}.

amount of the analyte in the first sampling compared to the second sampling of the same surface, the method is considered efficient [Boeniger et al., 2008].

Considering both gauze and twill wipes together, the first wipe in this evaluation usually removed the highest percentage of each flame retardant (median 48%; range: 1%–98%). The second wipe removed a median percent of 26% (range 1%–93%) of each flame retardant, and the third wipe removed a median of 23% (range: 0%–92%) of each flame retardant. The second and third wipes together removed a median of 52% (range: 2%–99%). However, an examination of the median percent removed by individual flame retardants showed that there were several cases in which the third wipe removed more than the first or second wipes. These median collection efficiencies of the total flame retardants appear to be comparable to collection efficiencies of wipe protocols that have been developed for other agents such as lead and polycyclic aromatic hydrocarbons [Ashley et al., 2009; Boeniger 2007; Boeniger et al., 2008]. The acceptable analytical variability for wipe flame retardant analysis in this study (60%–140%) was larger than that for other wipe sample analyses (80%–120%). The frequent presence of a high percentage (>50%) of flame retardants on the second and third hand wipes suggests that a single wipe is likely not sufficient to quantitatively characterize the extent of contamination on skin. This indicates that studies that have used only one wipe likely underestimated the quantity of some flame retardants on the skin surface. A higher log K_{ow} value may indicate a greater affinity for that flame retardant to adhere to the skin requiring several wipes to breakdown the natural skin-oils before the flame retardant could be removed and collected. We observed that for the flame retardants with log K_{ow} >7.5 removal levels by the third wipe exceeded the first wipe for 33% of the analytes and 55% of the analytes exceeded those detected in the second wipe. For flame retardants with log K_{ow} <7.5, the levels removed by the third wipe never exceeded the levels on the first wipe, and only 10% of the analytes exceeded those detected in the second wipe.

Our second objective was to compare two different hand wipe materials, gauze and twill. The majority of the studies we reviewed used gauze wipes [Allen et al., 2013; Carignan et al., 2013; Hoffman et al., 2015; Stapleton and Dodder, 2008; Liu et al., 2017]. Although we did not see a statistically significant difference between the two wipe materials, our sample size was small. The results of our pilot study still suggest that twill wipes could potentially be a better wipe material than gauze. Twill wipes removed a greater median percent in the first wipe sample, had less variability in the percent removed by the first wipe as indicated by the smaller interquartile range, and we didn't see evidence of a relationship between the total amount of flame retardant on the twill wipe and the percent removed. The twill wipe material was denser than the loosely woven gauze wipe, which may make transfer easier. Additionally, the gauze wipes were 3in by 3in while the twill were 4in by 4in therefore the twill wipes had a higher surface area than the gauze wipes.

There were several limitations to this study. For example, the third wipe was not analyzed at the same time as the first and second wipes, although we used surrogate corrected results in an attempt to control for this. In addition, the surrogate for certain organophosphate flame retardants (TCEP, TCIPP, TBP, TDCIPP, TPHP, and TCP) and TBBPA analysis in the third wipe was very low ($\leq$LOD to 16 μg), and therefore we determined that the analysis was suspect and results. In addition, we had a wide variability of results which could be from interpersonal sampling variation, such as participants wiping their hands differently, thereby removing the flame retardants from a previously un-wiped portion of the hand. Furthermore, differences in applied pressure and speed of wiping affect the quantity removed per wipe. In addition, we do not know how differences in skin moisture, the use of skin care products, handwashing, or the use of hand sanitizer affects the sampling results, nor did we collect data on the frequency or time (proximity to sample collection) of participant handwashing throughout the day. Similarly, we could not standardize exposures as would be done in an absolute dose study, therefore, it is possible that two employees working in the same department have different exposure levels resulting in different skin loadings. We also did not standardize the sample results with an estimate of hand surface area per sample. Although isopropyl alcohol is typically used as the wetting agent for hand wipes for flame retardants in much of the current scientific literature, only a single wipe sample has been collected [Watkins et al., 2011, 2013; Stapleton and Dodder, 2008; Hoffman et al., 2015]. In our study, the repeated wipes caused our participants' skin to become dry and irritated so we determined that three or more sequential wipes were not advisable while using isopropyl alcohol.

Hand wipes provide a direct measure of contaminants in the skin. Dermal sampling can help the researcher determine if hand-to-mouth or take-home exposures (i.e., carrying contaminants from the work to home) may be occurring. Dermal sampling can also determine the predominant types of flame retardants in the work environment. However, absolute quantitative values may be difficult to interpret and the precise number of sequential wipes required to fully quantitate the amount of flame retardant on an employee's skin is uncertain. The research community could greatly benefit from a laboratory based dosing study and a sequential wipe study using a variety of different wetting agents such as a lower percent of isopropyl alcohol (i.e. 70%) or corn oil so that repeat wipes can be collected and not irritate the skin. Regardless, this is the first study designed to evaluate the need for sequential wipes and to compare gauze and twill wipes for the sampling of flame retardants on skin.

5. Conclusion

Our study demonstrated that the first gauze or twill hand wipe typically removed the highest percent of flame retardants. However, the median percent removed by the first wipe was only 48% of the sum of three wipes, looking at gauze and twill together which means that a median of 52% of the flame retardants remained on the hands of the participant after the first wipe. In addition, a median of 23% of the total flame retardants remained on the third wipe, which may suggest that the participant may still have had flame retardants on their hands after the third wipe. As this was not an absolute dosing study, it can be assumed that these percentages for total flame retardants removed may be overestimates. We also found that the total dose on the skin surface and the lipophilicity of the flame retardant may impact the quantity removed with a single wipe sample. One wipe likely underestimates the level of skin contamination, and a series of wipes may be needed to more accurately quantify dermal exposure to the more lipophilic flame retardants. Lastly, our study indicated that twill wipes may be better than gauze, but more research is needed.

Disclaimer

The findings and conclusion in this report are those of the author(s) and do not necessarily represent the official position of NIOSH policy. Mention of trades names and or commercial products does not constitute endorsements or recommendations for use. The authors have no known conflicts of interest in conducting and reporting this research.

Acknowledgements

The authors would like to thank Bureau Veritas North America (now Maxxam) for sample analysis, and Rosa Key-Schwartz and Paula Fey-O'Connor for chemical analysis and quality control. This paper is contribution No. 3794 of the Virginia Institute of Marine Science, College of William & Mary. Diana Ceballos is a JPB Environmental Health Fellow. The JPB Foundation supports the JPB Environmental Health Fellowship Program, which is managed by the Harvard Chan School of Public Health. Diana Ceballos's time writing this manuscript was supported by grant NIH/NIEHS 2R25ES023635-04.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2018.12.027.

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