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Legacy and alternative flame retardants in house dust and hand wipes from South China

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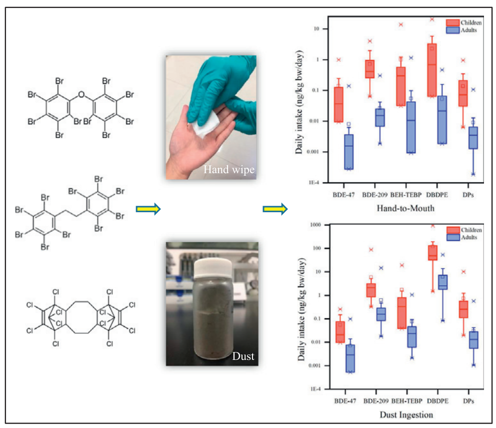
HIGHLIGHTS

- A wide range of flame retardants was determined in South China house dust and hand wipes from adults and children.
- BDE-47, BDE-209, DBDPE, BEH-TEBP, and DPs were frequently detected in hand wipes.
- Dust concentrations of BDE-47 and DPs significantly influenced their levels in hand wipes.
- Exposure via dust ingestion and hand-to-mouth contact was estimated for both children and adults.

GRAPHICAL ABSTRACT



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ABSTRACT

The present study investigated the occurrence of legacy and alternative halogenated flame retardants (FRs) in house dust ($n = 51$) from Guangzhou, South China and hand wipes collected from adults ($n = 51$) and children ($n = 31$). In addition to polybrominated diphenyl ether (PBDE) congeners (particularly BDE-209), several alternative FRs were also detected in >60% of dust samples, including decabromodiphenylethane (DBDPE; median: 4600 ng/g), bis(2-ethylhexyl)-3,4,5,6-tetrabromo-phthalate (BEH-TEBP; 43.9 ng/g), 1,2-bis(2,4,6-tribormophenoxy)ethane (BTBPE; 9.2 ng/g), pentabromotoluene (PBT; 10.1 ng/g), and *syn-* and *anti*-dechlorane plus (DPs, 24.5 ng/g). BDE-47, BDE-209, DBDPE, BEH-TEBP, and DPs were also frequently detected on hand wipes from children (median mass: 0.1–1.1 ng) and adults (0.1–1.2 ng). Linear regression models suggest that dust concentrations of BDE-47 and DPs had significant or marginally significant associations with their masses on children's ($10^{\beta} = 2.82$; 95% CI: 1.20, 6.64 and $10^{\beta} = 5.57$, 95% CI: 1.85, 16.75, respectively) and adults' hands ($10^{\beta} = 4.46$; 95% CI: 0.92, 21.58 and $10^{\beta} = 5.11$; 95% CI: 1.74, 14.96, respectively), whereas no association was observed for any other FRs. Most of the investigated demographic, environmental, or behavioral factors did not significantly influence the levels of halogenated FRs on human hands. Estimation of human exposure risks via hand-to-mouth contact and dust ingestion indicates that children are subjected to elevated exposure than adults, and the relative importance of the two pathways is chemical-specific.

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1. Introduction

After the phase-out of polybrominated diphenyl ether (PBDE) flame retardants (FRs) in North America and Europe, a number of alternative flame-retardant chemicals have been subjected to increased use to meet flammability standards. More than 75 brominated and chlorinated chemicals have been reported with commercial flame retardant applications (Covaci et al., 2011). These include 2-ethylhexyltetrabromobenzoate (EH-TBB) and bis(2-ethylhexyl)-3,4,5,6-tetrabromo-phthalate (BEH-TEBP) which are considered replacements for PentaBDEs, as well as 1,2-bis(2,4,6-tribormophenoxy) ethane (BTBPE) and decabromodiphenylethane (DBDPE) which are considered as a major replacement for OctaBDE and DecaBDE, respectively (Covaci et al., 2011; USEPA, 2013; Ma et al., 2012). Chlorinated flame retardants mainly include *syn*- and *anti*-dechlorane plus (DP) and other dechlorane-like chemicals, such as Dechlorane 601 (Dec-601), Dec-602, Dec-603, Dec-604, and Dec-604 Component B (Dec-604CB) (Sverko et al., 2011). These various brominated and chlorinated flame retardants possess diverse physicochemical properties and differ in environmental behavior and fate, thus representing different environmental and human health risks.

House dust has been frequently used to measure indoor chemical contamination and human exposure risks. Humans are exposed to dust-associated chemicals via incidental ingestion of dust particles, inhalation of resuspended dust, and dermal absorption (Whitehead et al., 2011). PBDEs have been reported to be ubiquitously present in indoor dust (de Wit et al., 2012; Harrad et al., 2010a; Harrad et al., 2010b; Harrard et al., 2016; Venier et al., 2016; Whitehead et al., 2011). Several studies reported the associations between dust-associated PBDEs and human internal exposure levels, indicating the contribution of dust ingestion to human exposure (Coakley et al., 2013; Vorkamp et al., 2011). For example, a New Zealand study reported that breast milk concentrations were correlated with floor dust concentrations for BDE-47, BDE-183, BDE-206, and BDE-209, and with mattress dust for BDE-47, BDE-153, BDE-154, and BDE-209 (Coakley et al., 2013). Associations between dust pentaBDE concentrations and human serum levels of free T4, total T3, estradiol, sex hormone binding globulin, or follicle stimulating hormones suggested influences of dust-associated PBDEs on human health (Johnson et al., 2013). In addition to PBDEs, alternative FRs were also investigated in house dust from different regions, but the studies varied in the types of investigated chemicals, partly due to the diversity of alternative FRs. Investigations of human exposure to alternative FRs in indoor environment also remain limited compared with numerous PBDE studies.

Our very recent study evaluated a suite of organophosphate esters (OPEs) in Guangzhou (South China) house dust and hand wipes from adults and children living in those investigated homes (Tan et al., 2018). Our findings revealed significant influence of dust on the levels of selected OPEs on children's hands (Tan et al., 2018). Hand wipe has been reported as a better predictor of human internal exposure to selected flame retardants (e.g., pentaBDEs, tris(1,3-dichloropropyl) phosphate or TDCIPP, and triphenyl phosphate or TPHP) than dust (Hoffman et al., 2015; Stapleton et al., 2012; Watkins et al., 2011). Evaluations based on hand wipes also suggest that hand-to-mouth contact represents an important pathway in addition to dust ingestion, particularly for children (Hoffman et al., 2015). As a follow-up study, the present work aimed to investigate a variety of legacy and alternative, halogenated FRs in Guangzhou house dust and associated exposure to both adults and children. Specific objectives were to: (1) measure the types and concentrations of halogenated FRs in dust and human hands; (2) explore any predictors of continuous halogenated FR levels on human hands; and (3) estimate exposure risks via dust ingestion and hand-to-mouth contact for adults and children.

2. Materials and methods

2.1. Chemicals and reagents

In addition to 20 PBDE congeners, a total of 33 alternative FRs were measured in the present study. They included 20 brominated substances, including 2,4,6-tribromophenyl allyl ether (ATE), BEH-TEBP, BTBPE, decabromodiphenylethane (DBDPE), EH-TBB, hexabromobenzene (HBBZ), hexachlorocyclopentadienyl-dibromocyclooctane (HCDBCO), pentabromobenzyl acrylate (PBBA), pentabromobenzyl bromide (PBBB), pentabromobenzene (PBBZ), pentabromoethyl benzene (PBEB), pentabromotoluene (PBT), 1,3,5-tribromobenzene (TBB), tetrabromo-*o*-chlorotoluene (TBCT), α- and β-1,2,5,6-tetrabromocyclooctane (TBCO), α-, β-, and γ-1,2-dibromo-4-(1,2-dibromoethyl)cyclohexane (TBECH), and 2,3,5,6-tetrabromo-*p*-xylene (TBX). Additional 13 dechlorane-related chemicals were also measured, including monodechlorinated DP (Cl_{11}-DP), didechlorinated DP (Cl_{10}-DP), chlordene plus (Cplus), DP mono adduct (DPMA), hexachlorocyclo-pentadiene (HCCP), hexachloro (phenyl)norbornene (HCPN), Dec-601, Dec-602, Dec-603, Dec-604, and Dec-604CB, and *syn*- and *anti*-DP. Surrogate standards included 4′-fluoro-2,3′,4,6-tetrabromodiphenyl ether (F-BDE69), 4′-Fluoro-2,3,3′,4,5,6-hexabromodiphenyl ether (F-BDE160), and 2,2′,3,3′,4,5,5′,6,6′-nonabromo-4′-chlorodiphenyl ether (4PC-BDE208), while 3'-Fluoro-2,2′,4,4′,5,6′-hexabromodiphenyl ether (F-BDE154) was used as the internal standard. Reference standards of the above mentioned chemicals were purchased from Wellington Laboratories (Guelph, ON, Canada) or AccuStandard (New Haven, CT).

2.2. Participant recruitment and sample collection

Our study recruited 51 families residing in the city of Guangzhou, South China via verbal spread and social media. Major recruitment criteria were introduced by our previous study (Tan et al., 2018). A total of 51 adults (one from each family) and 31 children aged between 1 and 5 years old (one from each of the 31 selected families) were requested to fill out a short questionnaire, collecting information on sex, age, height, weight, occupation (for adults only), hand washing frequency, hours per day staying at home, home size, and the number of electronic equipment in homes. Children completed the questions with the assistance from their parents. Summary of the study population characteristics is given in Table S1 (Supplementary material).

Dust was collected from living room and bedrooms with a commercial vacuum cleaner (Electrolux, ZMO1511, 1400 W) attached with a pre-cleaned, customized nylon bag with a pore size of approximately 25 μm. The nylon bag was detached following dust collection and wrapped with clean aluminum foil. Hand wipes were collected from adults or children by wiping both hands on the palm and back of each hand with pre-cleaned gauze pads. The participants were requested not to wash hands during at least 2 h prior to wipe sampling. The collected wipes were wrapped with pre-cleaned aluminum foils and stored in clean glass jars. Pre-cleaned gauze pads and sodium sulfate were also used as field blanks (one for every five homes) for wipe and dust collection, respectively. Sieved dust (through a 125-μm cloth sieve) and hand wipes, as well as field blanks, were stored at −20 °C.

2.3. Flame retardant residue analysis

Approximately 20–50 mg of dust was spiked with surrogate standards and extracted with 5 mL of a mixture of hexane and dichloromethane (HEX: DCM; 1:1, v/v) under sonication. The extraction was repeated three times (5 min each) and the combined extract was cleaned through a solid phase extraction (SPE) cartridge packed with 2-g silica sorbent (Isolute, Biotage Inc., Charlotte, NC). After the cartridge was pre-conditioned with 10 mL HEX and then loaded with the concentrated extract, it was washed with 3 mL of HEX and then eluted with 11 mL of HEX/DCM mixture (6:4, v/v). The latter fraction contains target FRs and was concentrated and spiked with FBDE-154. Extraction of hand wipes followed the same procedures described above with the only exception that only half of the combined extract was subjected to SPE cleanup.

Quantitative measurement of legacy and alternative FRs was conducted on an Agilent 7890B gas chromatography equipped with a 15-m DB-5HT column (0.25 mm i.d., 0.1 μm, J&W Scientific) and coupled to a 5977A single quadrupole mass analyzer (Agilent Technologies, Palo Alto, CA) in electro-capture negative ionization (ECNI) mode. The injector was operated in pulsed-splitless mode (held at 260 °C). The initial column temperature was set at 50 °C (held for 3 min) and then ramped to 300 °C at 8 °C/min (held for 10 min). Quantification was conducted based on each FR's characteristic ions under the selected ion monitoring (SIM) mode (Table S2).

2.4. Quality assurance and control

A dust composite was prepared by pooling dust samples from different sampling campaigns. Spiking tests were conducted by using this dust composite to demonstrate method validity. Approximately 50 ng each of the target FR analytes was spiked with dust composite (25 mg) and processed in five replicates with the method described above. Two dust composite samples were also processed without spiking target analytes. The recoveries of spiked FRs, after subtracting the original concentrations in dust composite (average values from two replicates), ranged from 75.8 $\pm$ 9.2% to 103.4 $\pm$ 5.9%. Pre-cleaned gauze pads ($n = 5$) were also spiked with 5–10 ng each of the target analytes and their recoveries from sample process ranged from 84.4 $\pm$ 5.6% to 95.2 $\pm$ 6.7% of the spiked values. Laboratory procedural blanks (one for every 10 samples) exhibited no contamination in the final extracts with the exception for BDE-47 which was detectable in approximately half of the blanks but with levels mostly below its limit of quantification (LOQ). Field blanks for dust sampling contained BDE-47 and BDE-209, but the levels in final extracts were below their LOQs, whereas in field blanks for wipe sampling only BDE-47 was detectable (<LOQ). Analysis of the National Institute of Standard Technology Standard Reference Material 2585 House dust revealed that the recoveries of PBDE congeners ranged from 87.5 $\pm$ 4.7% to 97.6 $\pm$ 4.3% of the reference values after adjustment with surrogate standards. An analyte's LOQ, defined as its response 10 times the standard deviation of the noise, ranged from 2 to 10 ng/g dry weight (dw) for dust analysis and 0.02–0.2 ng for hand wipe analysis. An analyte was considered non-detectable (nd) if its response is below the instrumental detection limit, i.e., a response three times the standard deviation of a noise.

2.5. Exposure estimation

Human exposure risks were evaluated via two approaches: dust ingestion and hand-to-mouth contact. The following equation was used to estimate daily FR exposure through dust ingestion (E_{DI}; ng/kg body weight/day) (Abdallah and Covaci, 2014; He et al., 2016):

$$E_{DI} = \frac{DIR \times C \times IEF}{BW} \quad (1)$$

where *C* is a FR's concentration in house dust (ng/g), *IEF* is the indoor exposure fraction (hours spent in homes within a day), *DIR* is the dust ingestion rate (g/day), and *BW* is body weight (kg).

Estimated daily exposure via hand-to-mouth contact (E_{HTM}; ng/kg body weight/day) was determined as (Stapleton et al., 2008):

$$E_{HTM} = \frac{M_{surf} \times TE \times SAC \times EF}{BW} \quad (2)$$

where M_{surf} is a chemical's mass on hands (ng), *SAC* is the proportion of the hand area contacted each time (%), and *TE* and *EF* represent the transfer efficiency (%; i.e., fraction of a chemical's mass transferred at each contact) and the frequency of contact during a day (day^{-1}), respectively.

2.6. Data analysis

Concentration data were corrected with the recoveries of corresponding surrogate standards, i.e., FBDE-69 (for analytes with a retention time earlier than BDE-85), 4PC-BDE208 (for BDE-209 and DBDPE), or FBDE-160 (for all other analytes). For measurements below LOQ for an analyte with a DF > 60%, a half LOQ was assigned for statistical analysis if its geometric standard deviation is greater than three; otherwise, a LOQ/$\sqrt{2}$ was assigned (Liu et al., 2018). Concentration data were determined for normal distributions with the Kolmogorov-Smirnov test. Logarithmical transformation was used to treat non-normally distributed data for statistical analyses. Paired-Samples *t*-Test was employed to determine the difference in FR levels between matched adult and children hand wipes from the same homes ($n = 31$ each). In addition to Spearman's correlation analyses of FR levels between hand wipes and house dust, linear regression models were also developed to explore predictors of continuous FR levels on hand wipes for adults and children separately. Except for age, all considered demographic and environmental factors were determined to be categorial variables and were dichotomized based on questionnaire data for linear regression models (Table S1). Dust concentrations were categorized into tertiles as predictors of FR levels on hand wipes. Statistical analyses were conducted with PASW Statistics 18.0 (IBM Inc.) with $\alpha = 0.05$ as the level of significance.

3. Results and discussion

3.1. Flame retardants in house dust

Among the 20 PBDE congeners screened, BDE-47, BDE-201, BDE-206, BDE-207, BDE-208, and BDE-209 had a detection frequency (DF) >60%. Concentrations of ΣPBDEs (including all detectable PBDE congeners) ranged from 118 to 27,980 ng/g (median: 520 ng/g) in Guangzhou house dust (Table 1). BDE-209 dominated the PBDE congener profile, constituting an average of 59 $\pm$ 21% of the total PBDEs. Concentrations of ΣPBDEs in Guangzhou house dust were generally within the range of concentrations reported in house dust worldwide (i.e., 50–9280 ng/g dw) (Peng et al., 2017).

DBDPE (DF $=$ 100%) dominated the FR composition profile (Fig. 1) and exhibited one order of magnitude greater concentrations (i.e., 150–96,400 ng/g; median 4600 ng/g) than PBDEs in the same house dust (Table 1). Concentrations of DBDPE in Guangzhou house dust were generally one to two orders of magnitude greater than the levels reported in house dust from most other countries (i.e., <10–2730 ng/g dw; Peng et al., 2017), suggesting extensive use of DBDPE in Chinese home products. Even higher DBDPE levels were reported in house dust from electronic waste recycling villages from China (i.e., 1160–26,300 ng/g) (Wang et al., 2010; Zheng et al., 2015). Among the variety of brominated FRs currently in use, DBDPE has the second highest demand in China, with an estimated production volume of 12,000 tons in 2006 alone and an estimated 80% annual increasing rate (Covaci et al., 2011; Xiao, 2006). Although DBDPE has limited bioavailability due to its great molecular weight and octanol-water partition coefficient (i.e., log $K_{ow} = 11.1$; Covaci et al., 2011), it exhibits an even greater affinity with dust particles than BDE-209, representing greater risks to humans via dust ingestion.

Additional alternative FRs frequently detected (i.e., DF ≥ 60%) in house dust include BEH-TEBP (median: 43.9 ng/g), BTBPE (9.2 ng/g), PBT (10.1 ng/g), and *syn-* and *anti*-DP (24.5 ng/g in combination) (Table 1). BEH-TEBP is a major component of commercial mixtures Firemaster 550, Firemaster BZ-54, and DP-45 (Ma et al., 2012). Along

Table 1
Concentrations (ng/g) or masses (ng) of legacy and alternative flame retardants in Guangzhou house dust and hand wipes from children and adult participants.

	Dust (n = 51)			Children's hand wipe (n = 31)			Adults' hand wipe (n = 51)		
	% Detect	Median	Range	% Detect	Median	Range	% Detect	Median	Range
BDE-47	71	5.5	nd[a]–180	74	0.1	nd–1.6	75	0.1	nd–7.5
BDE-201	82	14.8	<LOQ[b]–138	13	nd	nd–0.2	16	nd	nd–0.4
BDE-206	92	30.6	<LOQ–940	26	<LOQ	nd–0.9	31	<LOQ	nd–1.8
BDE-207	92	30.6	<LOQ–835	58	0.2	nd–1.3	55	0.2	nd–2.9
BDE-208	96	51.9	<LOQ–475	45	<LOQ	nd–1.1	49	<LOQ	nd–1.7
BDE-209	100	293	33.6–26,340	94	0.7	nd–6.3	86	0.8	nd–16.1
∑PBDEs[c]		521	118–27,980		1.2	nd–10.5		1.7	nd–24.0
BEH-TEBP	78	43.9	nd–1940	61	0.5	nd–21.7	71	0.6	nd–61.1
BTBPE	78	9.2	<LOQ–149	10	nd	nd–0.2	22	nd	nd–0.8
DBDPE	100	4600	153–96,410	74	1.1	nd–31.9	75	1.2	nd–24.7
EH-TBB	31	nd	nd–534	10	nd	nd–21.2	20	nd	nd–23.5
HBBZ	43	<LOQ	nd–85.2	35	<LOQ	nd–0.2	27	<LOQ	nd–0.2
PBT	69	10.1	nd–150	16	nd	nd–0.2	8	nd	nd–0.2
∑BFRs[d]		5028	226–96,670		1.3	nd–33.1		2.5	nd–71.2
syn-DP	78	5.3	<LOQ–216	68	0.02	nd–0.2	76	0.04	nd–1.5
anti-DP	98	19.4	<LOQ–834	97	0.1	nd–1.4	94	0.14	nd–5.3
∑DPs[e]		24.5	nd–1050		0.1	nd–1.5		0.2	nd–5.6

[a] nd = no detection.
[b] LOQ = limit of quantification.
[c] ∑PBDEs, including all detectable PBDE congeners.
[d] ∑BFRs, including all detectable brominated FRs excluding PBDEs.
[e] ∑DPs, including *syn*-DP and *anti*-DP.

with another Firemaster 550 component EH-TBB, BEH-TEBP is considered as a PentaBDE replacement, while BTBPE (marketed as FF-680) has been used to replace OctaBDE since 2005 (Hoh et al., 2005). EH-TBB was also detected in 31% of dust samples, but the median concentration was less than LOQ. PBT belongs to a family of brominated benzene FRs, which also includes ATE, HBBZ, PBBA, PBEB, PBBZ, TBB, TBCT, and TBX, but the latter chemicals were generally not detectable except for HBBZ (DF = 43.1%). Little is known about the exact production and applications of brominated benzene FRs (Venier et al., 2015), although some of them have been frequently detected by the Global Atmosphere Passive Sample (GAPS) network (Lee et al., 2016).

Syn- and *anti*-DP are the only dechlorane-like chemicals detected in house dust, with the combined concentrations (referred to as ΣDPs) ranging from nd to 1050 ng/g dw. The f_{anti} values (i.e., concentration ratio of *anti*-DP to ΣDPs) ranged from 0.43 to 1.0 (mean: 0.81) in house dust, comparable to the composition in technical DP mixtures where f_{anti} generally ranges from 0.65 to 0.75 (Qiu et al., 2007). The other chlorinated FRs were generally not detectable. This agrees with our previous report in house dust from another Chinese metropolitan region (Peng et al., 2017), as well as in most studies from other countries. These data may indicate that dechlorane-based chlorinated FRs other than DPs have not been subjected to broad applications in home products.

In addition to the FR chemicals discussed above, other screened alternative FRs were generally not detectable or detected at a DF < 50%. It should be noted that following the phase-out of PBDE mixtures, some of the alternative FRs have been subjected to increasing applications, likely resulting in increasing environmental releases and levels over time. Some alternative FRs (e.g., EH-TBB, BEH-TEBP, BTBPE, DBDPE, and dechlorane-related chemicals) are more and more frequently reported in not just indoor environment, but also in human bodies (Butt et al., 2014; Cequier et al., 2015; Hoffman et al., 2014; Zhou et al., 2014).

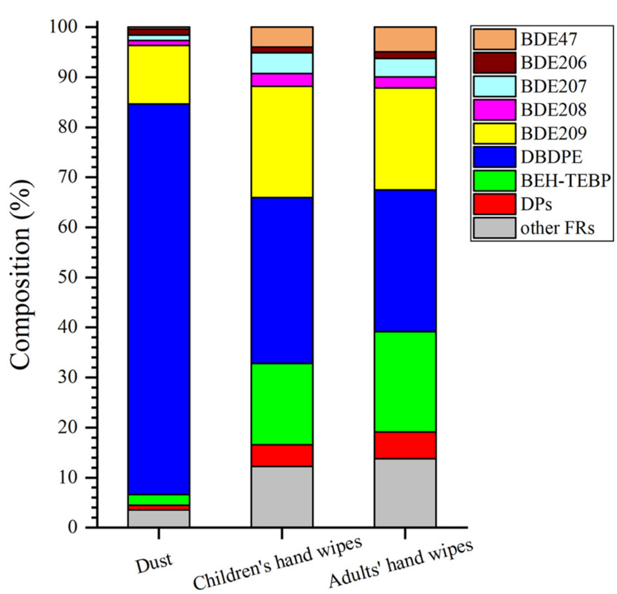

Fig. 1. Compositions of halogenated flame retardants in dust and hand wipes from children and adults.

3.2. Flame retardants on hand wipes

Hand wipe sampling provides an estimation of the amounts of contaminants present on hands. Flame retardant chemicals with a DF > 60% on hand wipes include BDE-47, BDE-209, DBDPE, BEH-TEBP, and DPs (including *syn*- and *anti*-DP), which had a median mass of 0.1, 0.7, 1.1, 0.5, and 0.1 ng on children's hand wipes, and 0.1, 0.8, 1.2, 0.6, and 0.2 ng on adults' hand wipes, respectively (Table 1). Similar to the pattern observed in dust, DBDPE and BDE-209 also dominated the FR chemical composition on hand wipes (Fig. 1). Proportions of BDE-47, BDE-209, BEH-TEBP, and DPs to the total FR levels on hand wipes were all significantly greater than the respective values in dust, whereas the proportion of DBDPE was significantly lower on hand wipes ($p <$ 0.05 in call cases).

No significant differences were observed for any of these substances between matched adults' and children's hand wipes. However, considering that children's hand surface areas are smaller than those of adults, the hand surface normalized levels of these detected FRs should be much higher for children than adults, likely representing a greater exposure risk for children. Additionally, no significant correlations were observed between matched adults' and children's hand wipe levels for any of the detected FRs. This suggests that the FRs present on adults' and children's hands may originate from different sources.

Using hand wipes to measure FR levels on human hands has been employed by several studies, but mostly focusing on PBDEs and OPEs (Hammel et al., 2016; Hoffman et al., 2015; Stapleton et al., 2012; Watkins et al., 2011; Xu et al., 2016). The U.S. studies revealed frequent detections of several PBDE congeners (e.g., BDE-28, -47, -66, -85, -99, -100, -153, -183, and -209) on hand wipes collected from multiple populations, with the reported median or geometric mean ΣPBDE mass ranging from 35 to 129 ng (Stapleton et al., 2008; Stapleton et al., 2012; Stapleton et al., 2014; Watkins et al., 2011). These levels were one to two orders of magnitude greater than the total mass of PBDEs on hand wipes from our study. This is due to the historically greater demands of PBDE mixtures, particularly PentaBDEs, in the U.S. than China. The North American market has consumed >95% of the world's PentaBDE production (Chen and Hale, 2010). Greater usage consequently resulted in higher levels of PBDEs on human hands from U.S. populations than the levels reported elsewhere. Fewer studies have reported the occurrence of alternative halogenated FRs on hand wipes. Stapleton et al. (2014) reported the detection of BEH-TEBP (GM = 2.5 ng; DF = 53%) and EH-TBB (GM = 4.1 ng; DF = 93%) on U.S. hand wipes, as well as hexabromocyclododecane diastereomers and tetrabromobisphenol A with much lower levels (i.e., 1.0 and 0.4 ng, respectively).

Overall, the investigations of FRs on human hands via hand wipe sampling remain limited. Although hand wipe levels of BDE-47, BDE-209, DBDPE, BEH-TEBP, and DPs were low for our adult and children participants, their frequent detections suggest high chances of human exposure through hand-to-mouth contact. Correlations between hand wipes and serum/urine concentrations for selected FRs also suggest that hand wipe has a potential as an alternative sampling approach for evaluating exposure without directly collecting human samples (Hoffman et al., 2015; Stapleton et al., 2012; Watkins et al., 2011). Therefore, hand wipe evaluations contribute valuable information to the elucidation of human exposure to indoor chemicals.

3.3. Predictors of FR levels on hand wipes

Linear regression models were employed to determine the predictors of continuous FR levels on hand wipes by including dust, demographic, and behavioral data. Only the FRs frequently detected on hand wipes, including BDE-47, BDE-209, DBDPE, BEH-TEBP, and DPs, were included in the models. The beta coefficients were exponentiated to produce the multiplicative change on hand wipe levels relative to the per-unit change for continuous variables (age only in our study) or the reference group for categorical variables (Hoffman et al., 2015).

Dust significantly influenced hand wipe levels only for selected FRs. Children participants with the highest dust concentrations (3rd tertile) of BDE-47 and ΣDPs in their homes averaged 2.82 times (95% CI: 1.20, 6.64) and 5.57 times (95% CI: 1.85, 16.75) the hand wipe levels compared with those with the lowest dust concentrations, respectively (Table 2; Fig. 2 for Spearman's correlation). No significant association was observed for BDE-209, DBDPE, or BEH-TEBP. Similarly, for adults only DPs exhibited a significant association between dust and hand wipe levels ($10^{\beta} = 5.11$, 95% CI: 1.74, 14.96), while a marginal association was observed for BDE-47 ($10^{\beta} = 4.46$, 95% CI: 0.92, 21.58). This indicated that sources other than house dust could substantially impact BDE-209, DBDPE, and BEH-TEBP levels on participants' hands. For example, frequent touch with FR-containing products may lead to the attachment of very small particles with hands or even the direct sorption of chemicals on skin lipids. Webster et al. (2009) reported that bromine detected in house dust was associated with a polymer/organic matrix. The small, bromine-containing particles are formed via abrasion or weathering processes and may be directly attached with human hands. In addition to house dust, outdoor dust could also influence the levels on hands. BDE-209 and DBDPE have been detected with greater abundances than other PBDE congeners in outdoor dust (Newton et al., 2015; Ding et al., 2016; Drage et al., 2016; Yu et al., 2012). A study on office environment also found that while a marginally significant association was observed between office dust and hand wipes for pentaBDEs, no association was observed for BDE-183 or BDE-209 (Watkins et al., 2011). Therefore, the influence of indoor dust on FR levels present on hands is chemical-specific. Using indoor dust alone to predict FR levels on hands may only be applicable to selected chemicals.

Hand washing frequency exhibits no significant influence on the levels of any FRs on children's or adults' hands (Table S3). This is different from our previous findings in the same populations, which indicated that hand washing frequency inversely influenced selected OPEs or the total OPEs in children's hand wipes, but the influence was much weaker for adults (Tan et al., 2018). The studied brominated and chlorinated FRs generally have greater lipophilicity than OPEs according to their K_{ow}; thus the former groups of FRs are better associated with skin lipids and less removable by hand washing. Watkins et al. (2011) also reported that hand washing frequency significantly influenced pentaBDE levels on office workers' hands, but no influence was observed for BDE-183 or BDE-209. Therefore, the influence of hand washing frequency on the levels of FRs on hands appears to be chemical-specific, likely determined by a variety of factors, such as lipophilicity, the sources and pathways from which a FR becomes associated with hands, and possibly other behavioral factors. These merit further elucidations.

None of the other considered demographic or environment factors, including age, sex, indoor temperature, humidity, the number of electronic equipment in homes, and dwelling size, was associated with hand wipe levels of target FRs (Table S3). The only exception is for BEH-TEBP, which was associated with dwelling size for children participants only ($10^{\beta} = 12.22$, 95% CI: 1.52, 98.17). However, the underlying factor is unclear.

It should be noted that our results may be confounded by other factors, such as the variations in hand areas between individual participants, particle size-dependent abundances of FRs in dust, and relatively small sample sizes. Normalization of hand wipe data with hand surface area may adjust for the variations in hand areas. However, we did not directly measure hand surface areas in the present study and using experimental models to estimate hand surface areas may bring additional variance to the analysis. Partially due to this consideration, several recent hand wipe studies also used FR masses without hand surface area normalization for data analysis and discussion (Hoffman et al.,

Table 2
Regression analyses for brominated flame retardants in dust as predictors of hand wipe levels.

Compounds	Low dust levels	Children's hand wipes				Adults' hand wipes			
		Mid dust levels		High dust levels		Mid dust levels		High dust levels	
		Coefficient[a] (95% CI)	p-Value	Coefficient (95% CI)	p-Value	Coefficient (95% CI)	p-Value	Coefficient (95% CI)	p-Value
BDE-47	Reference	1.22 (0.54–2.77)	0.61	2.82 (1.20–6.64)	0.02	2.90 (0.72–11.64)	0.13	4.46 (0.92–21.58)	0.06
BDE-209	Reference	1.45 (0.40–5.30)	0.56	0.94 (0.22–4.04)	0.93	1.63 (0.62–4.31)	0.32	1.12 (0.43–2.93)	0.81
BEH-TEBP	Reference	3.21 (0.25–40.55)	0.35	4.69 (0.43–51.64)	0.20	3.65 (0.35–38.02)	0.27	4.25 (0.46–39.08)	0.20
DBDPE	Reference	0.04 (0.01–0.94)	0.05	0.18 (0.01–3.95)	0.26	0.87 (0.25–3.07)	0.83	1.00 (0.27–3.67)	1.00
∑DPs	Reference	3.74 (1.15–12.16)	0.03	5.57 (1.85–16.75)	0.004	3.62 (1.42–9.23)	0.01	5.11 (1.74–14.96)	0.004

[a] Coefficient = Exponentiated beta coefficient, representing the multiplicative change on hand wipe levels relative to the reference group of dust concentrations.

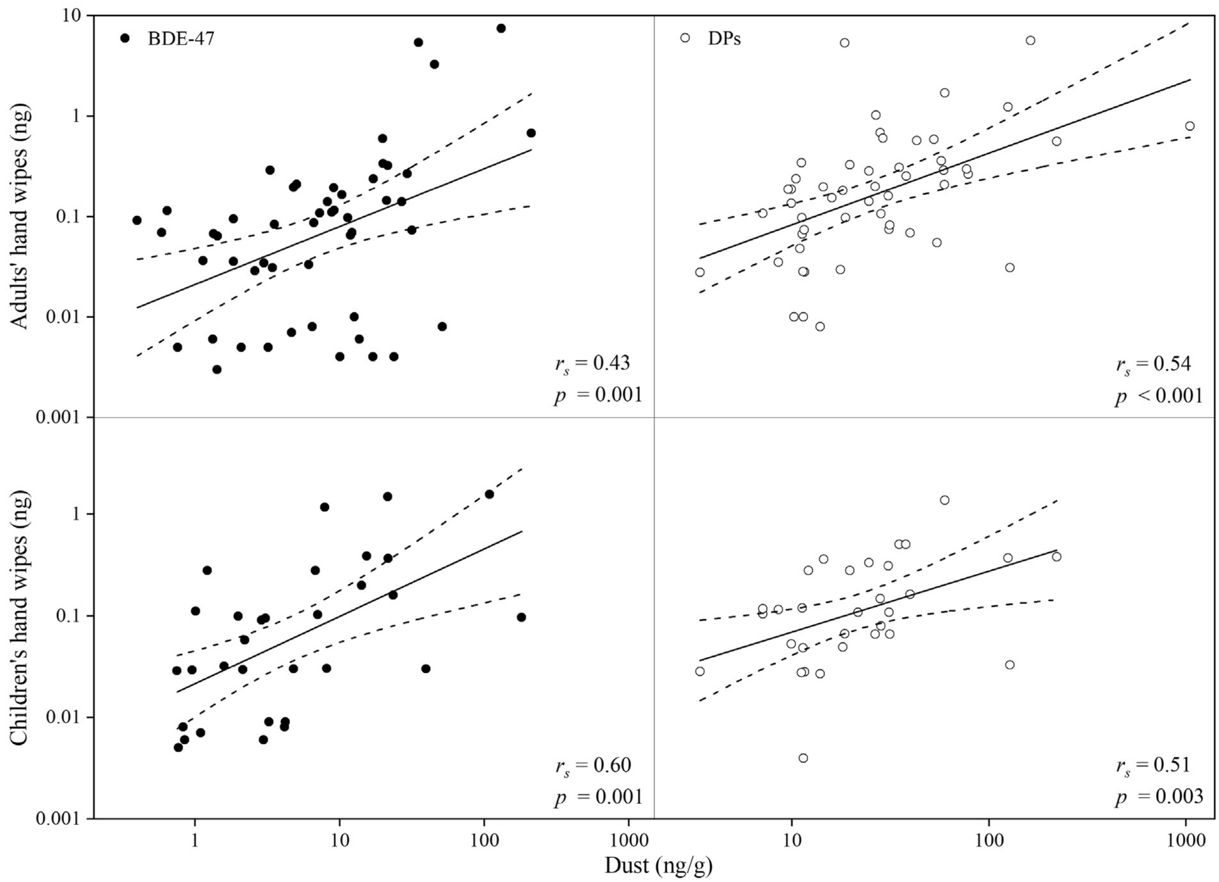

Fig. 2. Spearman's correlations between dust and hand wipes in the levels of BDE-47 and ΣDPs. Dashed lines represent the ±95% confidence bands.

2015; Hammel et al., 2016; Stapleton et al., 2014; Watkins et al., 2011). Additionally, the dust samples evaluated in the present study represent a holistic group of particles with sizes < 125 μm. We did not fraction them into smaller size groups. However, particle size-dependent abundances of FRs have been reported in indoor dust. For example, Cao et al. (2014) reported the increase of concentrations for BDE-209, DBDPE, and *anti*-DP along with the decrease of particle sizes (i.e., ranging from 900 to 2000 μm to ~7 ± 7 μm). Considering that the particles present on hands are usually very fine particles, exploration of particle size-dependent distributions may facilitate a better elucidation of dust influence on FR levels on hands. To address these limitations identified for the present study, our future expanded studies will take hand surface area normalization and particle size-dependent distribution into consideration.

3.4. Exposure estimation

Human exposure risks to FRs via dust ingestion and hand-to-mouth contact were estimated in the present study. Only BDE-47, BDE-209, BEH-TEBP, DBDPE, and DPs were included in the estimation of exposure via hand-to-mouth contact, while additional FRs detected with a DF > 60% in dust were also included in dust ingestion estimation. It should be noted that dust ingestion and hand-to-mouth contact are not completely independent. Dust ingestion represents total exposure through the ingestion of dust particles on the surfaces of furnitures or consumer products, floor dust, as well as dust present on hands. Hand-to-mouth contact represents one form of ingestion of contaminants associated with dust present on hands or absorbed to skin lipids (Stapleton et al., 2008; Stapleton et al., 2014).

We summarized from the questionnaires that the average body weights of children and adult populations in our study were 17 and 64 kg, respectively, and they spent an average of 83.3% and 66.7% of their time in homes, respectively (Table S4). For adult participants we estimated a median exposure rate via dust ingestion to be 0.1 ng/kg bw/day for ΣPBDEs, 1.0 for ΣBFRs (all brominated FRs excluding PBDEs), and 0.01 for ΣDPs (Table 3) under the average exposure scenarios (assuming an average DIR of 20 mg/kg), and 0.3, 2.6, and 0.01 ng/kg bw/day under the high exposure scenarios (assuming a high DIR of 50 mg/kg), respectively (Abdallah and Covaci, 2014; Ali et al., 2013). Children were subjected to elevated exposure, i.e., 1.3, 12.3, and 0.1 under the average exposure scenarios (assuming a DIR of 50 mg/kg), and 5.1, 49.3, and 0.2 ng/kg bw/day under the high exposure scenarios (assuming a DIR of 200 mg/kg), respectively. Exposure estimation for BDE-47, BDE-209, BEH-TEBP, DBDPE, and DPs is also summarized in Table 3. Greater exposure for children likely results from their higher dust ingestion rate, lower body weight, and more time spent in homes.

Hand-to-mouth contact results in comparable exposure to BDE-47, BEH-TEBP, or DPs when compared with dust ingestion, but the exposure estimation for BDE-209 and DBDPE was one to two orders of magnitude lower via the former pathway. For example, a median daily DBDPE exposure rate for children is determined to be 0.7 ng/kg bw/day via hand-to-mouth contact, in contrast with the estimated rate of 11.3 and 45.1 ng/g kg bw/day via dust ingestion under the average and high exposure scenarios, respectively. Similarly, for adults the DBDPE exposure rate via hand-to-mouth contact (i.e., 0.02 ng/kg bw/day) was also much lower than the rates via dust ingestion under the average and high exposure scenarios (i.e., 1.0 and 2.4 ng/kg bw/day, respectively). Our previous OPE study indicated that the rate of exposure to OPEs through hand-to-mouth transfer is approximately 50% greater than that through dust ingestion under the average exposure scenarios, but lower than the exposure rate under the high exposure scenarios (Tan et al., 2018). Thus the relative importance of hand-to-mouth

Table 3
Estimation of exposure from dust ingestion and hand-to-mouth contact.

	Children				Adults			
	Median	Mean	5th	95th	Median	Mean	5th	95th
Hand to mouth								
BDE-47	0.04	0.1	<0.01	0.8	<0.01	0.01	<0.01	0.04
BDE-209	0.4	0.7	<0.01	1.9	0.02	0.03	<0.01	0.1
DBDPE	0.7	2.2	<0.01	9.9	0.02	0.05	<0.01	0.2
BEH-TEBP	0.3	1.0	<0.01	3.8	0.01	0.05	<0.01	0.1
∑PBDEs[a]	0.8	1.5	0.1	5.0	0.03	0.1	<0.01	0.2
∑BFRs[b]	0.8	3.8	0.1	17.8	0.1	0.1	<0.01	0.4
∑DPs[c]	0.1	0.1	0.01	0.4	<0.01	0.01	<0.01	0.03
Dust ingestion (average exposure)								
BDE-47	0.01	0.03	<0.01	0.1	<0.01	<0.01	<0.01	0.01
BDE-209	0.7	2.8	0.2	5.7	0.1	0.2	0.01	0.5
DBDPE	11.3	27.3	1.2	81.0	1.0	2.3	0.10	6.9
BEH-TEBP	0.1	0.3	<0.01	1.4	0.01	0.03	<0.01	0.1
∑PBDEs	1.3	3.6	0.4	6.6	0.1	0.3	0.03	0.6
∑BFRs	12.3	27.8	1.3	81.2	1.1	2.4	0.1	6.9
∑DPs	0.1	0.1	0.02	0.4	0.01	0.01	<0.01	0.03
Dust ingestion (high exposure)								
BDE-47	0.05	0.1	<0.01	0.4	<0.01	0.01	<0.01	0.02
BDE-209	2.9	11.1	0.7	22.8	0.2	0.6	0.04	1.2
DBDPE	45.1	109.1	4.7	323.8	2.4	5.8	0.3	17.2
BEH-TEBP	0.4	1.4	<0.01	5.5	0.02	0.1	<0.01	0.3
∑PBDEs	5.1	14.4	1.6	26.6	0.3	0.8	0.1	1.4
∑BFRs	49.3	111.2	5.4	324.8	2.6	5.9	0.3	17.3
∑DPs	0.2	0.6	0.1	1.4	0.01	0.03	<0.01	0.1

[a] ∑PBDEs, including all detectable PBDE congeners.
[b] ∑BFRs, including all detectable brominated FRs excluding PBDEs.
[c] ∑DPs, including *syn*-DP and *anti*-DP.

contact compared with dust ingestion appears to be chemical specific, likely affected by a number of factors, including a chemical's relative abundance in dust or on hands, its physicochemical parameters (e.g., lipophilicity), and the influence of environmental or behavioral factors (e.g., hand washing).

Despite of the low exposure rates via hand-to-mouth contact for brominated and chlorinated FRs, this exposure pathway should not be overlooked. Previous studies have demonstrated that hand wipe is better than house dust in the prediction of internal exposure levels for selected FRs (Stapleton et al., 2012). For example, hand wipe levels were demonstrated to be associated with serum concentrations of pentaBDEs in both office workers and toddlers from two U.S. studies (Stapleton et al., 2012; Watkins et al., 2011). Although both hand wipes and dust exhibited a significant association with serum in the total levels of BDE-47, BDE-99, and BDE-100, the relationship was more significant for hand wipes (Stapleton et al., 2012). Hand wipe levels of TDCIPP and TPHP were also found to correlate with their metabolite concentrations in urine, but such correlations were not found between matched dust and urine (Hoffman et al., 2015). Previous studies also suggest that hand wipes not only represent exposure risks via hand-to-mouth contact, but also indicate potential dermal absorption (Hoffman et al., 2015; Phillips et al., 2018). However, for chemicals not frequently detectable on hands, other pathways (i.e., dust ingestion or inhalation) are more important to human internal exposure.

Given the co-existence of multiple exposure pathways (i.e., dust ingestion, hand-to-mouth contact, dermal contact, and inhalation), the understanding of the relative importance of different exposure pathways becomes very important to the elucidation of human exposure risks to indoor chemicals. It is very likely that the most important pathways are chemical specific and determined by a complexity of factors, such as a chemical's octanol-air partition coefficient (K_{oa}) and K_{ow}, personal behavior, age, or indoor environmental characteristics. For example, our previous OPE study found that indoor temperature inversely affected hand wipe levels of selected OPEs and such influence is more significant on adults than children (Tan et al., 2018). Additionally, hand washing exhibited greater influence on hand wipe levels of OPEs collected from children versus adults, suggesting an age factor (Tan et al., 2018). In addition to dust, air, and hand wipes, biological samples (e.g., urine or serum) should be investigated whenever possible as they provide a better elucidation of the relative importance of different exposure pathways, as well as the variety of influencing factors.

4. Conclusion

In this study we investigated legacy and alternative FRs in house dust from Guangzhou homes and hand wipes collected from adults and children. The results reveal a significant or marginally significant association between dust and hand wipe levels of BDE-47 and DPs, but no significant association is observed for any other FRs. Most of the considered demographic or environment factors exhibited no impact on hand wipe levels of target FRs. Exposure estimation indicates that hand-to-mouth contact results in comparable exposure with dust ingestion with respect to BDE-47, BEH-TEBP and DPs, but the latter pathway constitutes a more important contribution to human exposure to BDE-209 and DBDPE, as well as other FRs frequently detected in dust but not on hand wipes. Future studies are needed to better elucidate the chemical-specific relative importance of different exposure pathways and the influencing factors.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.11.369.

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