EXAMINING PREDICTORS OF EXPOSURE TO POLYBROMINATED DIPHENYL ETHERS (PBDEs) AMONG CHINESE UNIVERSITY STUDENTS

By

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs) are flame-retardant compounds common in furniture, textiles, plastics, and electronic items. Throughout the use and disposal of products, PBDEs may enter the indoor or outdoor environment. Many studies have shown that PBDEs cause adverse human health effects, including but not limited to disruptions in thyroid and estrogen hormones, changes in fertility, and developmental effects. In order to decrease human exposure to PBDEs, it is important to understand the fate and behavior of these compounds in the environment. Globally, usage patterns differ between geographical regions. Although fate and distribution of PBDEs has been well studied in the U.S. and Europe, much less is known about exposure in Asia, specifically for China. The purpose of this study was to document the levels of major commercial PBDE mixtures, and common replacement compounds (Firemaster® 550), in handwipes taken from a small population of Chinese students as an estimate of exposure potential. Additionally, PBDE levels were compared to differences in behavioral and lifestyle variables to see if any were significant predictors of exposure. It was hypothesized that time spent indoors, number of electronic items, hand washing frequency, and cleaning behaviors would influence the PBDE levels found on the hands. In each handwipe sample, at least one of the PBDE congeners or alternative compounds were detected, suggesting that the Chinese population is being exposed to

these compounds. Several behavioral and product variables were significantly correlated to exposure levels and others were suggestive of a relationship. For PentaBDE, floor cleaning method was a significant predictor of exposure (p=0.02). For DecaBDE, floor cleaning method and number of computers in the room were significant predictors (p=0.02, p=0.05). Other variables showed large differences in the mean flame retardant levels between groups, such as time spent indoors, transportation behavior, and time spent watching TV, but were not statistically significant. Due to the small sample size and non-normal distribution of PBDE levels in the study population, it is recommended that these variables be examined again with a larger sample size. To date, this is the first study to measure levels of PBDEs and alternative compounds in handwipes from a population in China.

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are a class of brominated flame retardant chemicals designed to reduce flammability of plastics, textiles, and other goods, for both commercial and residential use. They serve to delay ignition of materials by directly interfering with the combustion process[1]. Halogenated flame-retardant chemicals can be either reactive or additive in nature: either incorporated directly into the material or simply dissolved into the material[2]. The additive types, like PBDEs, tend to volatilize

[1] United States Environmental Protection Agency, An Exposure Assessment of Polybrominated Diphenyl

[2] Frank Rahman, et al., "Polybrominated diphenyl ether (PBDE) flame retardants," *Science of the Total Environment*, 275, (2010), 1.

and leach out of the material over time, leading to greater potential for environmental contamination and human exposure[3]. There are three main commercial mixtures used historically since production in 1976, PentaBDE, OctaBDE, and DecaBDE[4]. Penta-BDE is used in a wide variety of products including cushioning, textile and backing materials in furniture, and automobiles, as well as in epoxy resins in circuit boards, rubber, polyvinyl chloride (PVC) wire coatings, paints, polyurethane foam, lacquers, and adhesives[5]. OctaBDE is mostly utilized as an additive for acrylonitrile-butadiene-styrene plastics found in the casings and housings of televisions, computers, hair dryers, office equipment and automotive casings[6]. Additionally, DecaBDE is primarily used in electronic equipment such as TV's, audio/video, cell phones, remote controls, PCs, and monitors, cables but can also be found in nylon materials for sofas, chairs, and office furniture[7].

The most prominent homologue group, which differs by the amount of bromine substitution on the ring structure, explains the naming system used for commercial mixtures[8]. For PentaBDE, the commercial mixtures called DE-71™, DE-60™ or Bromkal 70-SDE ™, contain between 50-62% PentaBDE, 24-38% TetraBDE, and 4-12% HexaBDE[9] [10]. Specifically, it consists of 43% BDE-99, 28% BDE-47, 8% BDE-100, 6% BDE-153, 4%

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[3] Ibid, 1.
[4] USEPA, 2010, 2-2.
[5] USEPA, 2010, 2-7.
[6] Ibid, 2-7.
[7] Ibid, 2-8.
[8] Rahman, 2010, 3.
[9] USEPA, 2010, xvii.
[10] United States Environmental Protection Agency, Toxicological Review of 2,2',4,4',5-Pentabromodiphenyl ether (BDE-99) (Cas No. 60348-60-9), 2008, (Washington, D.C., U.S. EPA, 2008), 3.

BDE-154, and less than 1% of BDE-28, 33, 49, and 66 combined[11]. The Octa-BDE mixture, also called DE-79, contains the following congeners; BDE-183 (40%), BDE-197 (21%), BDE-203 (5-35%), BDE-196 (8%), BDE-207 (7%), BDE-208 (10%), BDE-153 (5-10%), and BDE-154 (1-5%)[12]. In the commercial Deca-BDE mixture, or DE-83R™ and Saytex 102E™, the main constituent (almost 97%) is BDE-209[13]. Thus, the distribution of congeners in biota and environmental compartments will largely depend on the source of the PBDEs, as different items contain different mixtures.

GLOBAL PRODUCTION & REGULATION

The Americas, Europe, and Asia represent 98% of the global demand for PBDEs[14] (Table 1.) The highest demand in the Americas is for DecaBDE (24,500 tons) and PentaBDE (7100 tons), 44 and 95 percent of the global total. In Europe, the highest tonnage (7,600 tons) is for DecaBDE. Furthermore, in Asia, DecaBDE and OctaBDE are the dominant mixtures, totaling 23,000 and 1,500 tons respectively. The demand for PentaBDE in Europe and Asia is only 4 percent of the global total.

Table 1. Global Market Demand for PBDE mixtures (tons) in 2001[15].

PBDE Mixture	Americas	%	Europe	%	Asia	%	Rest of World	%	Total

[11] Ibid, 3.
[12] USEPA, 2010, xvii.
[13] United States Environmental Protection Agency, Toxicological Review of Decabromodiphenyl Ether (BDE-209) (CAS No. 1163-19-5), 2008, (Washington, D.C., U.S. EPA, 2008), 3.
[14] Ronald A. Hites, "Polybrominated Diphenyl Ethers in the Environment and in People: A Meta-Analysis of Concentrations." *Environmental Science and Technology,* 38, (2004), 946.
[15] Hites, 2004, 946.

Penta	7100	95	150	2	150	2	100	1	7500
Octa	1500	40	610	16	1500	40	180	5	3790
Deca	24,500	44	7600	14	23000	41	1050	2	56150
Total	33100	49	8360	12	24650	37	1330	2	67440

As a direct result of concern regarding high global demand, ubiquity of PBDEs in the environment, and potential adverse health effects in animals and humans; there has been global and national legal regulation of the commonly used commercial mixtures. In 2001, the European Union (EU) placed a ban on both the PentaBDE and OctaBDE mixtures. Three years later, additional legislation banned the DecaBDE mixture[16]. The United States Environmental Protection Agency (USEPA) followed suit in 2004, convincing the only US manufacturer of PBDEs, the Great Lakes Chemical Corporation (Chemtura), to agree to a voluntary phase-out of PentaBDE and OctaBDE formulations by the end of 2005[17]. In 2009, producers and importers of DecaBDE agreed to discontinue the manufacture and sale of the chemical for most uses by 2012 and to stop using it altogether by the end of 2013[18]. Under the Toxic Substances Control Act (TSCA), the EPA proposed significant new use rules (SNURs) in 2006 to ensure that no new manufacture or imports of PBDEs occurs without being evaluated first[19]. Further amendments were made in 2012 that added provisions to the "new uses" to include

[16] United States Environmental Protection Agency, "DecaBDE Phase-out Initiative." *Existing Chemicals*, http://www.epa.gov/opptintr/existingchemicals/pubs/actionplans/deccadbe.html (Retrieved September 16, 2012).
[17] United States Environmental Protection Agency, "Polybrominated diphenyl ethers (PBDEs) Significant New Use Rules (SNUR)," *Existing Chemicals*, http://www.epa.gov/oppt/existingchemicals/pubs/qanda.html. (Retrieved January 12, 2013).
[18] USEPA, 2012, 4.
[19] Ibid, 4.

processing of Penta and Octa; manufacture, import, and processing of Deca; and manufacture, import, and processing of any item containing PBDEs.

There has been some regulation of PBDEs in China, including phase-out of PentaBDE and OctaBDE mixtures back in 2004[20]. The DecaBDE mixture continues to be used in China, mainly in electronic appliances and will be used in the U.S. until the end of this year. Despite regulation of new production of some PBDE mixtures in Europe, Canada, the US, and China, PBDEs will undoubtedly continue to be released into the environment as old products continue to be used, are discarded, or recycled.

Due to the phase out of some PBDE mixtures, replacement compounds have been introduced into the market. PentaBDE for use in furniture has been replaced by several compounds: TDCPP (tris(1,3-dichloroisopropyl)), Firemaster® 550, TPP (triphenyl phosphate), and TBPP (tris(4-butylphenyl)phosphate)[21]. Firemaster® 550 contains two main components; TBB (2,3,4,5-tetrabromo-ethylhexylbenzoate) and TBPH (2,3,4,5-tetrabromo-bis(2-ethylhexyl)phthalate[22]. The signature pattern of usage of Firemaster® 550 is the finding of TBB and TBPH compounds in a 4 to 1 ratio[23]. It is used in specific applications such as insulation for wiring and cables, backing in carpets, in wall coverings, coated fabrics, adhesives, and is even added to PVC and neoprene.[24].

[20] Ibid, 4.

[21] Heather, M. Stapleton, et al., "Novel and High Volume Use Flame Retardants in US Couches Reflective of the 2005 PentaBDE Phase Out," *Environmental Science and Technology*, 46, (2012), 13432.

[22] Jonathan S. Bearr, et al, "Accumulation and DNA Damage in Fathead Minnows (*Pimephales Promelas*) Exposed to 2 Brominated Flame Retardant Mixtures, Firemaster 550 and Firemaster BZ-54." *Environmental Toxicology and Chemistry*, 29, (2010): 722.

[23] Heather M. Stapleton, et al., "Alternate and New Brominated Flame Retardants Detected in U.S. House Dust," *Environmental Science & Technology,* 42, (2008), 6910.

[24] Adrian Covaci, et al., "Novel brominated flame retardants: A review of their analysis environmental fate and behavior," *Environment International,* 37, (2011), 536.

There are other sources for TBPH as well, such as in a product called DP-45[25]. The total U.S. production of TBPH between the years 1990 and 2006 totaled between 450 to 4500 tons per year[26].

CHEMICAL PROPERTIES

There are 209 different congeners or structures for PBDEs, each with its own unique chemical properties (Figure 1). The congener number, i.e. level of bromination impacts each structures behavior in the environment and in biota. The various chemical parameters governing these compounds can be seen in Table 2. Generally, PBDEs are lipophilic (fat-loving) and hydrophobic (water-hating)[27] The lipophilic behavior of PBDEs means that these compounds are likely to be found concentrated in fatty tissues of biota or to be sorbed onto sediments or organic materials such as dust or soil rather than in water. These behaviors are demonstrated by the log K_{ow} or log K_{oc} values. Additionally, their vapor pressures are highly variable; congeners with lower bromination are more likely to be found in the vapor phase than the higher brominated congeners, as these compounds are lower in molecular weight. The structures of the new use chemicals TBB and TPBH contain both rings and bromine atoms, much like the PBDEs (Figure 2). Due to the similar sources and chemical properties of the new use chemicals (i.e. TBB/TBPH), it is assumed that they will behave comparably in the environment.

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[25] Ibid, 536.
[26] Ibid, 536.
[27] USEPA, 2010, 2-2.

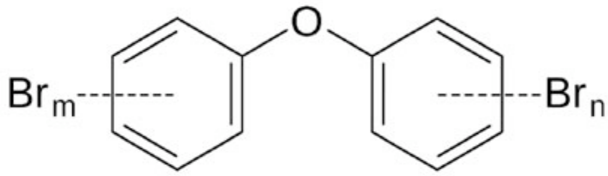

Figure 1. General chemical structure of a polybrominated diphenyl ether (PBDE)[28].

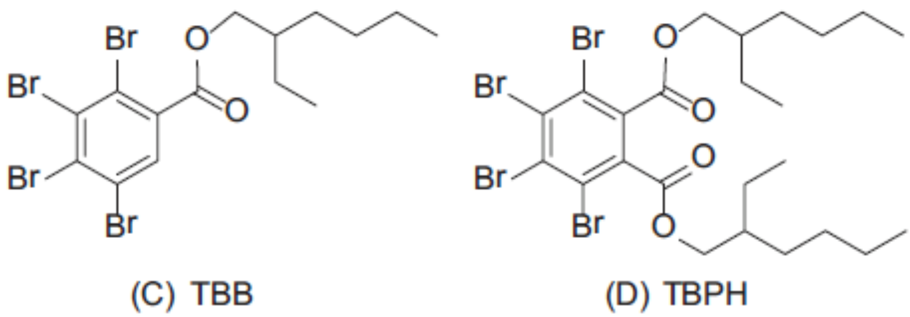

Figure 2. Chemical structure of compounds in Firemaster® 550; TBB and TBPH[29].

[28] Ministry of the Environment (2013). "Appendix B Polybrominated Diphenyl Ether (PBDE) structure and physical properties." from http://www.mfe.govt.nz/publications/hazardous/investigation-of-brominated-flame-retardants/html/page11.html.
[29] Heather B. Patisaul, et al., "Accumulation and Endocrine Disrupting Effects of Flame Retardants Mixture Firemaster 550 in Rats: An Explanatory Assessment," *Journal of Biochemical and Molecular Toxicology*, 27, (2013): 125.

Table 2. Chemical Properties of PBDEs[30]

CAS #	Penta-BDE 32534-81-9 Octa-BDE 32536-52-0 Deca-BDE 1163-19-5
Physical Description at Room Temperature	Pale yellow liquid or white powder
Molecular Weight (g/mol)	564-959.2 (Deca)
Water Solubility at 25 °C (ug/L)	1
Boiling Point (°C)	>300 to >400
Melting Point (°C)	85-306
Vapor Pressure at 25 °C (mmHg)	2.2×10^{-7} to 9×10^{-10}
Octanol-Water Partition Coefficient (K_{ow})	5.7 to 8.27
Soil organic C-Water Partition Coefficient (K_{oc})	4.89 to 6.80
Henry's Law Constant (atm m^3/mol)	7.5×10^{-8} to 1.2×10^{-5}

ADVERSE HEALTH EFFECTS

Although many of the health effects of PBDEs in humans are still not well understood, many adverse impacts have been found. As is the case with a number of persistent organic pollutants, health impacts for adults tend to be different than for children or fetuses. The effects on the endocrine system, however, are important for all life stages.

[30] United States Environmental Protection Agency, Technical Fact Sheet-Polybrominated Diphenyl Ethers (PBDEs) and Polybrominated Diphenyls (PBBs)", *Solid Waste and Emergency Response (5106P),* 2012, 2.

Like many other endocrine disrupting chemicals, PBDEs may be estrogenic[31]. By mimicking hormones in the body, they disrupt growth, development, reproduction, and behavior. Additionally, there is a structural analogy of PBDEs to the thyroid hormone T_4, resulting in interference with thyroid metabolism and transport.

Several studies indicate that there are neurodevelopmental effects following exposure PBDEs in fetuses and children. The primary route of exposure is via maternal transfer, either through the placenta in utero or from breast milk after birth. As expected, this causes infants to have higher body burdens of PBDEs, largely due to this transfer[32]. The concentrations found in the maternal and placental plasma were significantly correlated[33]. Furthermore, in utero exposure to PBDEs has been linked to adverse neurodevelopmental effects, such as lowered IQ and decreased performance on mental tests[34]. These adverse effects were detected at both early ages and as the children continued to age. In another study, postnatal exposure in four year olds to BDE-47 was significantly associated with poor social competence and increased risk of certain ADHD symptoms[35]. In pregnant women, increased concentrations of PBDEs measured in serum were linked to changes in fertility, leading to longer pregnancy times[36]. In

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[31] Michael Martin, et al., "An Asian quandry: where have all of the PBDEs gone?," *Marine Pollution Bulletin,* 49, (2004), 379.
[32] Leisa-Maree Toms, et al., "Higher Accumulation of Polybrominated Diphenyl Ethers in Infants Than in Adults." *Environmental Science and Technology*, 42, (2008), 7510.
[33] Marie Frederiksen, et al., "Polybrominated diphenyl ethers in paired samples of maternal and umbilical cord blood plasma and associations with house dust in a Danish cohort," *International Journal of Hygiene and Environmental Health,* 213, (2010), 233.
[34] Julie B. Herbstman, et al., "Prenatal Exposure to PBDEs and Neurodevelopment," *Environmental Health Perspectives,* 118, (2010), 716.
[35] Mireia, Gascon, et al., "Effects of pre and postnatal exposure to low levels of polybromodiphenyl ethers on neurodevelopment and thyroid hormone levels at 4 years of age," *Environment International*, 37, (2011): 605.
[36] Kim, G. Harley, et al., "PBDE Concentrations in Women's Serum and Fecundability." *Environmental Health Perspectives*, 118 (2010), 702.

men, increased exposure to PBDEs led to increased thyroglobulin antibodies, an indication of autoimmune thyroiditis and Grave's disease[37]. It also led to an increase of the thyroid hormone T_4. For these reasons, exposure to PBDEs is of concern for human health.

In contrast, little is known about the human health effects of components of Firemaster® 550. Recent animals studies indicate that these compounds could disrupt thyroid hormone levels, increase onset of puberty, cause changes in glucose levels, as well as being a potential obesogen[38]. Additionally, a dietary exposure study in fathead minnows showed reparable liver DNA damage and a potential to be bioavailable and accumulate in tissues[39]. Clearly, much more research is needed to understand the health impacts of these new compounds entering the environment.

FATE IN THE ENVIRONMENT

The potential for contamination of PBDEs to the environment can occur at various stages of product life cycle including during the "production, use, recycling, and disposal in domestic waste, landfill, and incineration of products containing PBDEs", making quantifying contribution from individual sources complicated[40]. During production, the main route of environmental release of Deca-BDE is through stack

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[37] Mary E. Turyk, et al.,"Hormone Disruption by PBDEs in Adult Male Sport Fish Consumers." *Environmental Health Perspectives*, 116, (2008), 1638.
[38] Patisaul, 2013, 132.
[39] Bearr, 2010, 728.
[40] Shinsuke, Tanabe, et al., "Brominated flame retardants in the environment of Asia-Pacific: an overview of spatial and temporal trends," *Journal of Environmental Monitoring*, 10 (2007), 188.

emissions[41]. Studies have shown that off-gassing, or volatilization of PBDEs from products within the indoor environment is also important[42]. Throughout the lifetime of the products, the PBDEs enter the indoor work or home environment by being released through "volatilization from polyurethane foam or abrasion from hard plastics"[43]. After release, they may enter the air, water, soil, and sediments in the outdoor environment or the air and dust within the indoor environment.

EXPOSURE ROUTES

No one particular exposure route has been cited to be the most important for humans globally, but tend to differ based on country or region. Dust is considered the major pathway for PBDE ingestion in the United States, while diet may be important for Europeans[44]. People living and working near 'e-waste' sites are likely to mainly be exposed to PBDEs via air emissions and during the breakdown of electronic equipment and plastics. The importance of different routes can vary depending on a person's age, lifestyle, behavior, and even the country in which they reside.

Diet

Diet plays an important role in the exposure of humans to PBDEs, although some foods may be more highly contaminated than others. Many studies have pointed to fish and

[41] USEPA, 2010, 2-12.
[42] USEPA, 2010, 2-12.
[43] Tanabe,et al., 2007, 195.
[44] Laurence Roosens, "Factors influencing concentrations of polybrominated diphenyl ethers (PBDEs) in Students from Antwerp, Belgium," *Environmental Science and Technology*, 42 (2009), 3535.

shellfish as being the major sources of dietary PBDEs, potentially due to bioaccumulation in marine food webs or aquatic sources of the flame-retardants[45]. However, factors such as age will also contribute to changes in dietary habits, and thus ingestion rates of PBDEs, especially for items such as meats and dairy products[46]. Another study found that in the U.S., red meat and poultry is a key dietary source of PBDEs[47]. Additionally, the levels of PBDEs between people classified as vegetarians versus omnivores were significant. As was mentioned earlier, PBDEs are hydrophobic and tend to accumulate in the fats within tissues. Therefore, food items like red meat, dairy, and seafood products with high lipid content usually contain higher levels of PBDEs.

Inhalation/Ingestion

Humans are in constant contact with numerous items containing PBDEs; items such as couches, chairs, carpet padding, electronic items (televisions, phones, computers), textiles, and mattresses[48]. They can even be found in automobiles, in the stereo, navigation system, and foam dashboard [49]. The major sources for human exposure to PBDEs can be either internal or external in origin[50]. Key routes of external exposure identified are ingestion, inhalation, and dermal absorption. Ingestion sources include

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45 Sumin Na, et al.,"Dietary Assessment of human exposure to PBDEs in South Korea." *Chemosphere,* 90, (2013), 1740.
46 Ibid. 1740.
47 Alicia J. Fraser, et al.,"Diet Contributes Significantly to the Body Burden of PBDEs in the General U.S. Population," *Environmental Health Perspectives*, 117, (2009), 1524.
48 Betts, 2008, A202.
49 Ibid, A202.
50 Marie Fredericksen, et al., "Human Internal and external exposure to PBDEs- A review of levels and sources," *International Journal of Hygiene and Environmental Health*, 212 (2009),111.

diet, especially foods high in fat (i.e. fish, meat, oils) and dust[51]. Unintentional ingestion of these chemicals, either from touching contaminated dust or consumer products, is mainly an issue of hand-to-mouth behavior[52]. Adults may smoke, bite their nails, or eat foods with their hands that lead to ingestion of these chemicals. Additionally, exposure via inhalation can come from PBDEs in indoor air, either in the gas or particle phase, or from outdoor air[53]. Dermal absorption is the direct uptake of the chemicals through the skin from consumer products or dust, but its importance is negligible. Taking into account all of the various routes of exposure in order to quantify risk levels is a complicated task.

Handwipes as a Measure of Exposure

As contact with products and ingestion of indoor dust are important human exposure routes, hand to mouth behaviors are likely to play a role. Therefore, several studies have utilized handwipes as a measure of this exposure [54-55.] Handwipes are collected by wiping the participants hand with a gauze pad soaked in alcohol in order to remove any PBDE residues present there at the time of collection. In Stapleton, et al., 2012, there were significant associations between PBDE levels measured in the handwipes and levels found in the serum, especially for BDE congeners 47, 99, and 100. The handwipes

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[51] Ibid, 2009, 112.
[52] Betts, 2008, A207.
[53] Frederickson, et al., 2009.
[54] Heather M. Stapleton, et al, "Serum PBDEs in a North Carolina Toddler Cohort: Associations with Handwipes, House Dust, and Socioeconomic Variables," *Environmental Health Perspectives*, 120, (2012), 1049-1054.
[55] Deborah J. Watkins, et al., "Exposure to PBDE's in the office environment: evaluating the relationships between dust, handwipes, and serum", *Environmental Health Perspectives*, 119, (2011): 1247.

were a better representation of exposure than measuring levels in the dust, because unlike dust they measured human exposure in multiple 'microenvironments'. In Watkins, et al. 2010, the relationship between handwipe levels, dust, and blood serum were examined and positive associations were found between all three variables.

Predictors of Exposure

It has been suggested that lifestyle differences between individuals may influence the importance of various exposure routes[56]. For one, differences in usage patterns and regulation between continents and geographical regions can lead to differential exposure to PBDEs. For example, years of residence in the United States was found to be an important predictor of PBDE exposure, with the more years residing in the U.S. the higher the levels of PBDEs [57]. Personal behaviors can influence exposure as well. As was discussed before, hand-to-mouth behaviors may be important exposure routes for PBDEs. For example, "smoking, nail biting, and eating oily finger foods such as French fries, nuts, and sandwiches with unwashed hands" can all lead to unintentional ingestion of PBDEs on hands[58]. Additionally, time spent in the indoor environment and in contact with products containing PBDEs is likely to increase exposure. In the case of exposure to dust, children (esp. toddlers) are crawling and playing on the floor, where the dust is likely to be accumulating. Therefore, the specific behaviors of these children may be influencing levels of exposure. Even socioeconomic status may influence exposure. In one study, a mother's education level and breastfeeding behavior

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[56] Tanabe, et al, 2007, 195.
[57] Harley, et al., 2010, 701.
[58] Betts, 2008, A207.

influenced the PBDE levels her child was exposed to[59]. Diet also influences exposure, as different foods contain varying levels of PBDEs, so personal food choices will play a role. More research is necessary to investigate these variables further and to understand which behaviors are most important for predicting exposure to PBDEs.

EXAMINING PREDICTORS OF EXPOSURE IN CHINESE STUDENTS

PBDEs IN CHINA

There is a high demand and national production of PBDEs within China as well as an enormous influx of these chemicals from other countries in the form of e-waste. PBDEs have contaminated every environmental compartment of China; the air, soil, sediments, and water. Lesser information is known, however, about the distribution and impacts on human health within the country.

Production and Usage

Asia is an important producer and consumer of halogenated flame-retardants, contributing 37% of the global demand for PBDEs prior to the phase-out of some of the commercial mixtures starting in 2004 [60]. Given strict regulation of PBDEs in the US, Canada, and Europe, the Asian-Pacific region is now the largest market for these

[59] Stapleton, et al., 2012, 1052.
[60] Tanabe, et al., 2007, 189.

compounds[61]. Furthermore, China will represent 50% of the global demand for flame-retardant chemicals by 2014. According to data gathered in 2007, Chinese domestic production of brominated flame-retardants was increasing at a rate of 8 percent annually from its 2007 level of 10,000 tons[62]. China is rare in that it locally produces PBDEs, in fact its domestic production of DecaBDE between 2000-2004 increased from 10,000 to 25,000 tons[63].

E-waste Recycling

South China may among the most contaminated areas in the world for PBDEs [64]. This is due, in part, to the presence of multiple large electronic waste cities and to budding electronics manufacturing industries[65]. The largest of these e-waste recycling areas is found in a town called Guiyu, located in Shantou in Guangdong Province. Many of the electronic waste activities are located in Southern China and may contribute to exposure in people who live near these areas[66]. The contribution of PBDEs from electronic goods is monumental; 1.7 million tons of "e-waste" were produced in 2006[67]. This amount is expected to increase over the next 11 years to almost 5.4 million tons[68].

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[61] Kun, Ni, et al., "Polybrominated diphenyl ethers (PBDEs) in China: Policies and recommendations for sound management of plastics from electronic wastes," *Journal of Environmental Management*, 115, (2013), 114-123.
[62] Tanabe, et al., 2007, 192.
[63] Agus Sudaryanto, et al., "Levels and congener specific profiles of PBDEs in human breast milk from China: Implication on exposure sources and pathways," *Chemosphere,* 73, (2008), 1661.
[64] Jian-Yang Guo, et al., "Polybrominated Diphenyl Ethers in Seafood Products of South China," *Journal of Agricultural and Food Chemistry,* 55, (2007), 9152.
[65] Ibid, 9152.
[66] Jing Zheng, et al.,"Levels and sources of brominated flame-retardants inhuman hair from urban, e-waste, and rural areas in South China," *Environmental Pollution,* 159, (2001), 3706-3713.
[67] Ni, et al, 2013, 115.
[68] Ibid, 2013, 115.

In Guangdong Province alone, approximately 145 million electronic devices are recycled, thought to contain 2.61 x 10^8 kg of PBDEs total[69]. The sources for these electronic good are from Europe and the United States and other outside sources. Unfortunately, it is common for developed countries to export electronic goods to Asia for disposal, mainly due to "low labor costs, lax environmental regulations, and lack of broad international regulations"[70]. This growing input of PBDEs to the environment in China is in stark contrast to the United States, where a voluntary phase-out of PentaBDE and OctaBDE are already in place and will extend to DecaBDE by the end of this year. Clearly, this influx of PBDEs is an important source to the Chinese environment and subsequent release will be key for human health outcomes.

Exposure

PBDEs are ubiquitous in the Chinese environment, having been found in marine mammals, fish, and sediments as well as in human adipose, breast milk and other human tissues[71]. PBDEs were found in breast milk from women in two Chinese cities, Nanjiang and Zhoushan. Nanjiang is a city of 6 million people and large textile, electronics, and automobile industries while Zhoushan has a population of 1 million with shipping, food, and pharmaceutical industries[72]. Furthermore, they have been found in high levels in the atmosphere, with e-waste facilities likely being a significant

[69] Martin, et al., 2004, 375.
[70] Ibid, 2004, 375.
[71] Robin, J. Law, "Levels and trends of HBCD and BDE's in the European and Asian environments, with some information for other BFR's,"*Chemosphere,* 73, (2008), 223-241.
[72] Sudaryanto, 2008, 1662.

contributor to this pollution[73]. A few studies have measured PBDE levels in dust in China. One such study found that BDE-209 made up almost 97% of the total amount of PBDEs measured in the dust[74].TBB and TBPH have even been found in porpoises and dolphins in Hong Kong[75].

Unlike the Northern hemisphere, however, the sources, fate, and distribution of PBDEs in China are not currently well characterized. China did not mandate the usage of PBDEs in the manufacture of furniture like the United States, notably in polyurethane foam, so furniture is not anticipated to be a significant source. More likely, the production of electronic goods within the country and degradation of these products over time will be the main source of PBDEs, represented by higher levels of DecaBDE in the samples.

PURPOSE OF THE STUDY

The purpose of this study was to provide more information about the levels of PBDEs and their replacement compounds in the Chinese environment. By measuring levels found on the hands from a small student population in China, the main PBDE mixtures that Chinese people are exposed to and in what levels were determined. Additionally, this study sought to examine the factors influencing exposure of PBDEs in mainland China populations. It also seeks to document the concentration of PBDEs found on the

[73]Ni, et al, 2013, 152.
[74] Laiguo Chen, et al., "Human Exposure to PBDEs via House Dust Ingestion in Guangzhou, South China," *Environmental Contaminant Toxicology*, 60, (2011), 556.
[75] James, C. W. Lam,"Temporal Trends of Hexabromocyclododecanes (HBCDs) and Polybrominated Diphenyl Ethers (PBDEs) and Detection of Two Novel Flame Retardants in Marine Mammals from Hong Kong, South China," *Environmental Science & Technology,* 43, (2009), 6944.

hands of Chinese students, in order to see which PBDE mixtures are being used and can be found in the Chinese environment. Furthermore, the relationships between home characteristics, behavior, and use of electronic items as they contribute to concentrations on hands and to exposure are explored. To my knowledge, this will be the first study documenting potential exposure of Chinese university students to PBDEs and measuring concentrations of PBDEs in handwipes from the Chinese population.

MAIN OBJECTIVES

1. Measure concentrations of PBDEs and replacement compounds in hand wipe samples from a population students at Shantou University;
2. Determine whether differences in behavior and lifestyle can explain variation in concentrations measured on the hands;
3. Compare concentrations of PBDEs measured in handwipes from this study with values reported elsewhere.

MATERIALS AND METHODS

Recruitment of Study Participants

Participants from Shantou University were recruited based on the criteria that the individual lived in university housing. Recruitment was performed by a small group of Shantou University students who walked around several campus dorms seeking subjects for the study. Most participants were graduate-level students between the ages of 20 and 30. Permission was granted by the Department of Biology at Shantou University.

All subjects were given a consent form, translated into Chinese, which informed them about the purpose of the study, the method of sample collection, and information about confidentiality. All participants signed the consent form prior to sample collection. Additionally, each participant was asked to fill out a questionnaire regarding personal information about diet, transportation usage, hand to mouth behaviors, and ownership of electronic equipment. The purpose of the questionnaire was to provide relevant information regarding potential routes of contact with PBDEs. A handwipe and dust sample was collected from each participant.

Site Description

Shantou University is located in Shantou, a city on the eastern coast of Guangdong Province in the People's Republic of China (Figure 3). It is located approximately only 24 miles from Guiyu, a major site of electronic waste disposal. It is a key coastal city for commercial, importing, and exporting activities[76]. The total area of the city is 2, 046 km^2 (707 sq mi) and has a population of 5,271,100[77]. It is an important region for trade and manufacturing, the main industries being canning, textiles, lithography, plastic, and

[76] People's Government of Shantou City, "Shantou.", *Website of Shantou Government,* http://english.shantou.gov.cn. (Retrieved 01/20/13).
[77] Ibid.

toys[78]. Shantou University has approximately 20,000 students and covers an area of 1.28 km^2.

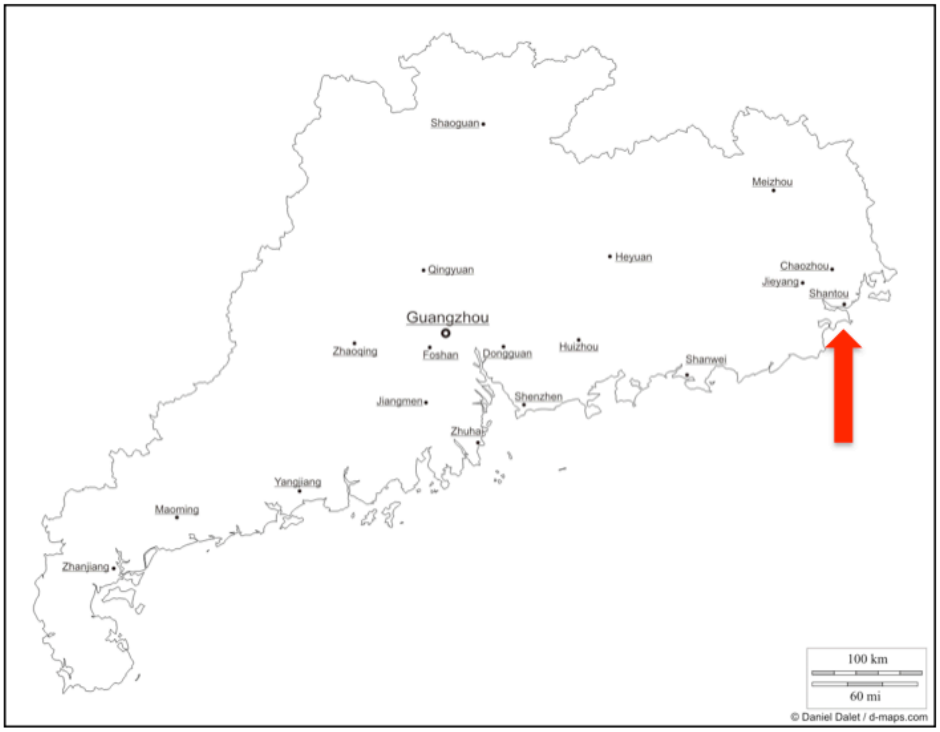

Figure 3. Map of Guangdong Province, People's Republic of China[79]

Dust Collection

Dust samples were collected from twenty-five participant's dorm rooms at Shantou University in Shantou, China on July 28th and 29th of 2012. Additionally, dust samples were collected from three different lab locations within Shantou University, where

[78] TriVista ,"Shantou," *Understanding China,* http://understand-china.com/?province=shantou. (Retrieved 1/20/13).
[79] http://www.rfa.org/english/news/china/liantang-11282012153054.html/china-guangdong-shantou-map-400.jpg

many of the participants worked. A Whatman cellulose thimble (Catalog number 2800288) dimensions 28mm x 80 mm, was secured into the Philips AirStar vaccum cleaner with bag, model no: FC822/01. The entire surface area, approximately 10 ft by 10 ft, of the dorm room was vacuumed. The cellulose thimble containing the dust was wrapped securely in foil and placed in a plastic bag. Field blank samples were taken by cleaning a section of floor with methanol, then wiping it dry with paper towels. Next, sodium sulfate was sprinkled on the area and vacuumed into a clean cellulose thimble. Samples were stored and secured in the same method as previously described. All dust samples were stored at -20C until analysis.

Handwipe Collection

Handwipe samples were collected from the same twenty-five participants at Shantou University. To collect handwipe samples, sterile *Cancare* brand gauze pads, 5 cm by 5 cm, were soaked in ~ 4 ml of reagent grade isopropyl alcohol. The subject's palmar and dorsal surfaces of the hand, as well as the sides of the fingers and palm were wiped with the isopropyl soaked gauze pad. The gauze was placed in a glass vial, wrapped in foil, and secured in a plastic bag. The samples were stored at room temperature for shipment between Shantou, China and Hong Kong, Hong Kong SAR and then again to Durham, North Carolina, where they were stored at -20 C until further analysis.

Chemical Analysis of Handwipes

The methods for extraction and chemical analysis for the handwipe samples followed protocol reported previously (Stapleton et al. 2008). For extraction, the gauze pads were spiked with 100 uL a recovery standard mixture containing the following; F-BDE 69, C^{13}-BDE 209, *d*-TDCPP (Tris (1,3-dichloro-2-propyl) phosphate), and *d*-TPP (triphenyl phosphate). The first extraction step was performed using Soxhlets with a 50:50 (v:v) mixture of hexane/DCM (dichloromethane) for at least 12 hours overnight. Following extraction, samples were reduced to approximately 1 ml volume via nitrogen gas using a TurboVAP machine. This step was followed by a clean up using Florosil solid phase extraction (SPE). For the first fraction, the SPE cartridges were first rinsed with 5 ml MeOH and 3 mL of hexane before samples are loaded. The samples were rinsed with approximately 2 ml of hexane. Then, PBDEs were eluted from the SPE with 4-5 ml of hexane. A second fraction containing organophosphate flame-retardants (OPFRs) was collected by elution with 10 ml of ethyl acetate, but will not be discussed in this paper.

Analysis was performed using gas chromatograph-mass spectrometry (GC/MS) in electron capture negative ionization mode. For quality assurance, several field blank hand-wipe samples were analyzed to evaluate background contamination.

Statistical Analysis

Statistical analysis was performed using Microsoft Excel. Data was first examined for normality using a Shapiro Wilk test and data was log transformed for further statistical analyses. The correlation coefficient, r, was measured using linear regression to determine whether BDE-47 and BDE-99 and TBB and TBPH concentrations were correlated. A simple t-test was run for each chemical group (PentaBDE, DecaBDE, Firemaster® 550 (TBB/TBPH), and TBPH) for each category of the following explanatory variables; transportation variables (bus, ferry, and car), time spent indoors and outdoors, cleaning behaviors (cleaning frequency, method of floor cleaning, method of furniture cleaning, electronics information (number of electronic items, age of computer/television, number of hours spent using computer/television). Each explanatory variable was divided into two or three categories based on 'high' and 'low' values for each in order to determine a difference in the means between the two categories. The relative difference between the means of the groups, reported as a percentage, was calculated by dividing the difference of the means by the average of the means. The relative difference represents the percent difference between the means of the two groups.

RESULTS

Population Characteristics

Table 3 summarizes the characteristics of the population of Shantou University students involved in this study. Most participants were male (80%) and graduate level (92%) students between the ages of 23 and 30. Most participants were in the normal weight range for their height, with only one underweight student and three overweight students. All participants resided in almost identical living conditions. Each dorm room had identical room dimensions, furniture type, and had tiled floors. Furthermore, every room lacked an air conditioning unit.

Table 3. Summary of Sample Population Characteristics (n=25)

Characteristic	n (%)
Sex	
Male	20 (80)
Female	5 (20)
Age	
<18	0 (0)
18-22	0 (0)
23-26	11 (44)
27-30	14 (56)
>30	0 (0)
BMI*	
Underweight	1 (4)
Normal	21(84)
Overweight	3 (12)
Education Level	
Undergraduate	2 (8)
Graduate	23 (92)

*BMI-Body Mass Index

PBDEs and Alternative Compounds in Handwipes

Each handwipe sample was tested for the following BDE congeners 17, 25, 28, 33, 47, 49, 66, 71, 75 85, 99, 100, 116, 119, 138, 155, 156, 181, 183, 190, 191, 200, 203, 205, 206, and 209. At least one of the PBDE congeners was found in each of the twenty-five handwipe samples. Certain congeners were not detected at levels above the method detection limit, including 30, 85, 155, 156, 190, 200, 203, 205, and 206. The largest contributors to the burden of PBDE in the handwipes (>36% of samples) were BDEs 28, 33, 47, 99, and 209. Both TBB and TBPH were detected in all of the handwipe samples. Other congeners such as BDE 49, 66, 75, 100, 153, and 183 were detected in the samples, but at a much lower frequency (Table 4). Some congeners were not included in the statistical analysis because of potential interference with another similar compound, such as BDE 75 and 191. The PBDE replacements TBB and TBPH were also frequently detected (>40% of samples). The commercial mixture PentaBDE is represented by BDE congeners 28, 33, 47, 99, 100, 153. The commercial mixture DecaBDE is represented by BDE congener 209. The new use replacement mixture called Firemaster® 550 is represented by TBB and TBPH. The compound TBPH is also used alone for statistical analysis due to the fact that it is used separately as a flame-retardant.

The widest range of levels found in the handwipes was for TBPH, between 1.54 and 326.8 ng. The highest level found for a BDE congener was for BDE 209, found in concentrations between 1.84 and 10.56 ng. The PentaBDE congeners, although present in all of the samples, were much lower than the concentrations of BDE 209, between 0.45 and 1.80 ng. Furthermore, the levels of the PBDE replacement compounds (TBB/TBPH) are found in much higher levels than the all the PBDE congeners besides BDE 209.

Table 4. Levels of PBDEs and Replacement Compounds in Handwipes

Congener	% Detectable	Median (ng)	Range (ng)
BDE 28,33	48	0.03	<0.05-0.32
BDE 47	44	0.26	<0.62-1.34
BDE 75	52	0.06	<0.12-0.21
BDE 99	36	0.16	<0.32-5.18
BDE 100	24	0.14	<0.13-0.19
BDE 153	12	0	<0.11-0.85

BDE 183	20	0	<0.07-4.98
BDE 191	44	0	<0.1-5.01
BDE 209	68	2.14	<1.84-10.56
ΣPentaBDE	43	0.87	<0.45-1.80
TBB*	48	0.35	<0.72-7.89
TBPH*	68	2.32	<1.54-326.8

ΣPentaBDE represents sum of BDE congeners 28,33,47,99, 100, 153.
*TBB/TBPH are PBDE replacements
*Congeners highlighted in gray were used in subsequent statistical analysis

Analysis of Normality

The data was examined for normal distribution by performing a Shapiro-Wilks test on each congener data. Furthermore, histograms were analyzed comparing the normal and log transformed data for each congener and congener group (PentaBDE, DecaBDE, Firemaster550, TBB, and TBPH). Log transforming the individual congeners did not appear to make the distribution much more normal for PBDEs, while log transformation did help for TBB and TBPH. Alternately, when the congeners were analyzed on a group basis, log transformation did appear to make the distribution more normal. Although the population was not normally distributed, the logarithmically transformed data were used in further statistical analysis.

Linear Regression

In order to determine whether or not the different congeners were correlated, an indication that they originate from the same source, a simple linear regression was performed. The BDE congeners 47 and 99 were significantly correlated ($R = 0.17$; p

=0.05). This suggests that BDE 47 and 99 may be coming from a similar source, such as from PentaBDE. Additionally, the compounds TBB and TBPH were highly correlated (R= 0.95). This correlation was statistically significant ($p < 0.01$). This suggests that TBB and TBPH have a similar source, such as the commercial mixture Firemaster® 550.

Description of Explanatory Variables

Due to the small sample size, each of the explanatory variables was divided into categories to provide a stronger statistical analysis. Table 5 shows the categories for each explanatory variable and what that category is describing.

Table 5. Description of Explanatory Variable Categories

Explanatory Variable	Category	Description
Hand washing	Low High	3-4 times per day > 4 times per day
Cleaning Frequency	Low High	1-2 times per week >3 times per week
Bus	Low High	1-2 hours per week >3 hours per week
Car	Low High	0 hours per week 1-2 hours per week
Indoors	Low High	5-10 hrs/day >10 hrs/day
Outdoors	Low High	<1 hr/day 1-5 hrs/day
# of Computers	Low High	≤1 >1
# of TVs	Low	0

	High	1
Time on Computer	Low High	≤5 hours >5 hours
Time on TV	Low High	0 hours >0 hours
Age of Computer	New Old	≤3 years >3 years

Analysis of Variance between Groups

The explanatory variables were split into different categories, summarized by Table 5.

Table 6 shows the results of the t-tests run between each mixture or compound (PentaBDE, DecaBDE, Firemaster® 550, and TBPH) and each explanatory variable.

Table 6. Differences in variation for PentaBDE, DecaBDE, Firemaster® 550, and TBPH between explanatory variable groups.

Explanatory Variable	Description			PentaBDE		DecaBDE		Firemaster550		TBPH	
	Category	*%*		*RD*	*p*	*RD*	*p*	*RD*	*p*	*RD*	*p*
Cleaning											
Hand washing	Low	28		42%	0.57	6%	0.9	33%	0.45	58%	0.33
	High	72									
Frequency	Low	84		38%	0.45	32%	0.52	17%	0.78	28%	0.74
	High	16									
Floor Method	Broom	28	Broom vs. Both	65%	0.02**	105%	0.02**	69%	0.21	106%	0.15
	Mop	36									
	Both	36	Broom vs. Mop	69%	0.07	27%	0.5	46%	0.27	35%	0.6
Transportation											
Bus	Low	56		52%	0.10	6%	0.88	3%	0.94	13%	0.84
	High	44									
Car	Low	68		28%	0.45	90%	0.06	53%	0.27	78%	0.27
	High	32									
Environment											
Indoor	Low	32		57%	0.19	3%	0.94	58%	0.14	80%	0.12
	High	68									
Outdoor	Low	12		40%	0.09	29%	0.68	38%	0.49	54%	0.44
	High	88									

Electronics											
# of Computers	Low	84		N/A	N/A	77%	0.05**	55%	0.23	78%	0.17
	High	16		N/A	N/A						
# of TVs	Low	68		N/A	N/A	49%	0.22	23%	0.59	13%	0.83
	High	32		N/A	N/A						
Time on Computer	Low	20		N/A	N/A	42%	0.34	60%	0.33	97%	0.3
	High	80		N/A	N/A						
Time on TV	Low	64		N/A	N/A	60%	0.11	6%	0.88	9%	0.88
	High	36		N/A	N/A						
Age of Computer	New	64		N/A	N/A	14%	0.72	7%	0.87	16%	0.79
	Old	26		N/A	N/A						

** indicates a significant p-value <0.05
%: Percentage of population
RD: Relative Difference (%)
p: p-value

DISCUSSION

Correlation between Congeners

The BDE congeners 47 and 99 were significantly correlated. It is likely that these congeners come from a common source, such as the PentaBDE mixture. Additionally, the compounds TBB and TBPH were significantly correlated. Similarly, it is likely that these two compounds originated from the same source, possibly the Firemaster550® mixture.

Concentrations in the Handwipes

At least one PBDE congener or alternative compound was found in each of the handwipe samples. The range of DecaBDE (BDE 209) in the handwipe samples is between 1.84-10.56 ng, much higher than the range for the PentaBDE mixture, 0.45 and

1.80 ng. The minimum value for DecaBDE is four times higher than for PentaBDE. The maximum value for DecaBDE is almost six times higher than for PentaBDE. Additionally, the median value found in the handwipes for DecaBDE (2.14 ng) is two times higher than the median value for PentaBDE (0.87 ng). This data indicates that there may be more sources for DecaBDE in the Chinese environment than for PentaBDE. As was mentioned earlier, PentaBDE was used in very small quantities prior to phase out in China, where as DecaBDE continues to be used and produced within the country. This aligns with the hypothesis that the DecaBDE mixture, represented by BDE 209, was predicted in higher concentrations in China than the congeners represented by the PentaBDE mixture.

Both TBB and TBPH were found at much higher concentrations than PBDEs in the handwipes. The range of concentrations measured for TBPH, between 1.54 and 326.8, were much higher than for TBB, between 0.72 and 7.89. The median concentration for TBPH was almost seven times higher than the median concentration for TBB. The minimum concentration measured for TBPH was two times higher than for TBB. Furthermore, the maximum level measured for TBPH was almost 50 times higher than the maximum level for TBB. Although both compounds are significantly correlated, indicating a common source, the values for TBPH are much higher. This may be evidence that there is another mixture containing TBPH, or that TBPH itself is being used in higher levels in China. Information regarding the amount of production or usage of TBB or TBPH in China is not currently available.

Predictors of Exposure

Hand washing

There were two categories for hand washing behavior; the 'high' group (>4 times per day) and the 'low' group (3-4 times per day). The high hand washing group represented 72% of the sample population and the low group represented 38% of the population. It is important to note that all participants were frequent hand washers given that no participant checked the categories for 0 times per day, and 1-2 times per day available the survey. Every participant washed his or her hands at least 3 times per day, if not more. Although hand washing behavior was not a good predictor of concentration of flame-retardants in this study, the p-values for Firemaster® 550 and TBPH were smaller than for the PBDE mixtures. This may point to hand washing as an important predictor of TBB or TBPH exposure, and also for PBDEs, but a larger sample size is needed.

Cleaning Behavior

For cleaning behavior, cleaning frequency and floor-cleaning method were examined. The cleaning frequency variable was divided into the 'low' (1-2 times per week) and 'high' (>3 times per week). The sample population that represented the 'low' category was 84% and was 16% for the 'high' category. Although the differences in means between the two groups were not significant for any flame-retardant group, the p-values for the BDE mixtures were smaller than for Firemaster® 550 or TBPH. Indoor dust may be more important for exposure to PBDEs than for the alternative compounds TBB and TBPH.

The floor-cleaning variable was split into three categories; whether the participant used a broom, a mop, or both a broom and a mop to clean their dorm room, known as 'broom', 'mop', or 'both'. The percentage of the population representing these categories was 28%, 36%, and 26%, respectively. For PentaBDE, the difference between the 'broom' group and the 'both' group was highly significant (p-value=0.02) with a 65% difference in the means (Figure 4). Also, the difference between the 'broom' group and the 'mop' group was suggestive, but not significant (p-value=0.07). Furthermore, the difference between the 'broom' and 'both' category for DecaBDE was significant (p-value=0.02) (Figure 5). These data suggest that floor dust may be a significant source for both PentaBDE and DecaBDE and that method of cleaning is important. It advocates that the use of both a broom and a mop in cleaning the floor results in lower dust levels and thus reduces the concentrations of PBDEs found on the hands. The differences between the groups for Firemaster® 550 and TBPH were not statistically significant; however, there was a 69% and 109% difference in the means for the 'broom' compared to 'both' group. With a larger sample size, the significance of floor cleaning and these alternative compounds may be revealed.

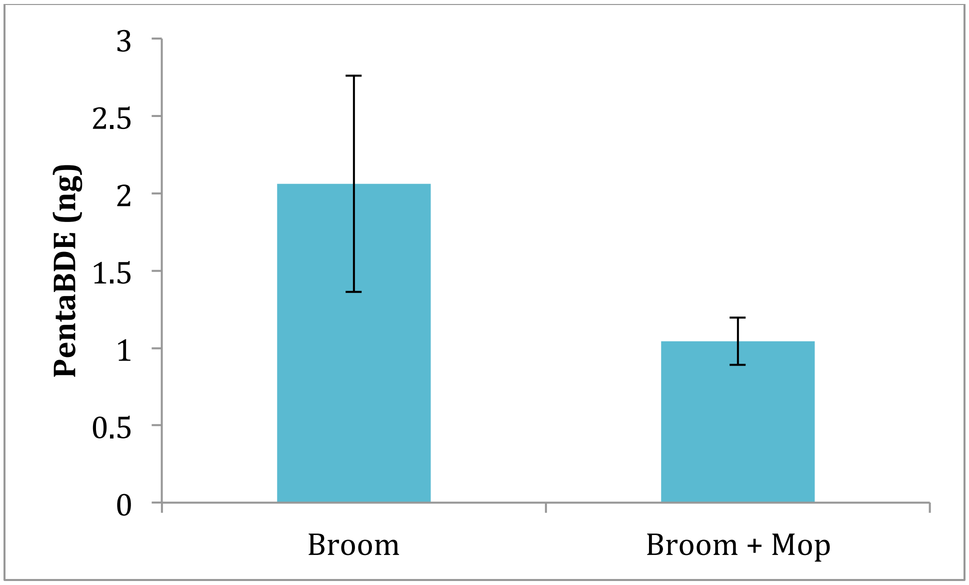

Figure 4. Comparison of PentaBDE levels on handwipes between students using a broom versus those reported using both a broom and mop.

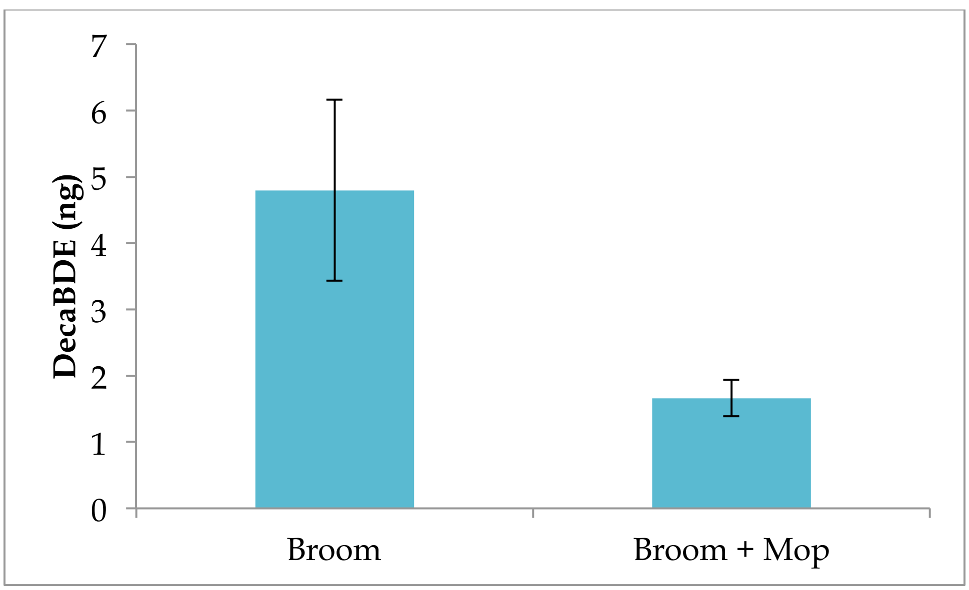

Figure 5. Comparison between DecaBDE levels on handwipes between students reported using a broom versus those reporting using both a broom and mop to clean.

Transportation

Each participant was asked how much time was spent per week using transportation methods such as a bus or car. Approximately 56% of the sample population demonstrated 'low' bus riding behavior (1-2 hours per week) and 44% were 'high' (>3 hours per week), while 68% demonstrated 'low' car riding (0 hours per week) and 32% 'high' (1-2 hours per week). From these values, it is clear that the sample student population rides the bus more hours a week than they do in a car per week. For bus riding, the 52% difference between the 'high' and 'low' groups was a suggestive predictor of exposure for PentaBDE (p-value=0.1) (Figure 6). For car riding, the 90% difference between the 'low' and 'high' groups for DecaBDE was also a suggestive predictor of exposure (p-value=0.06) (Figure 7).

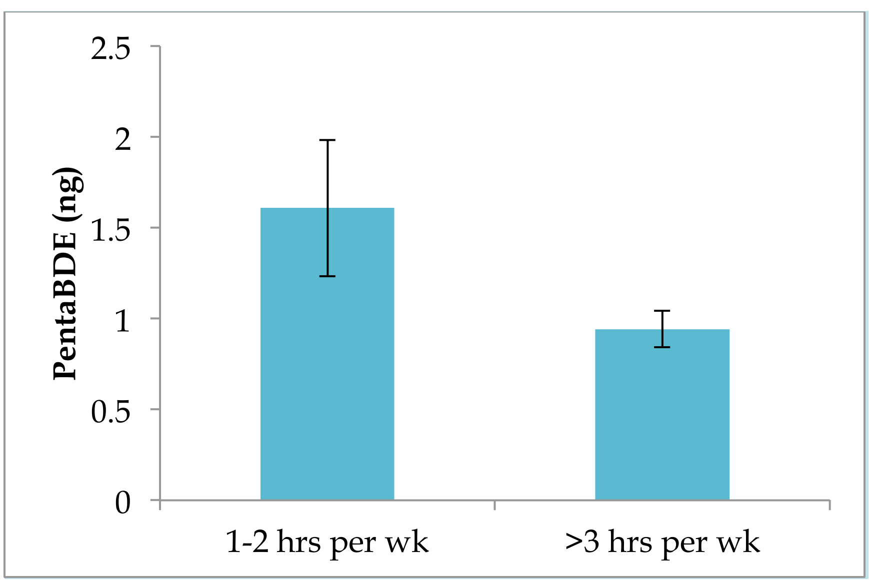

Figure 6. Comparison between PentaBDE levels on handwipes in individuals who reported spending 1-2 or >3 hrs per week in a car.

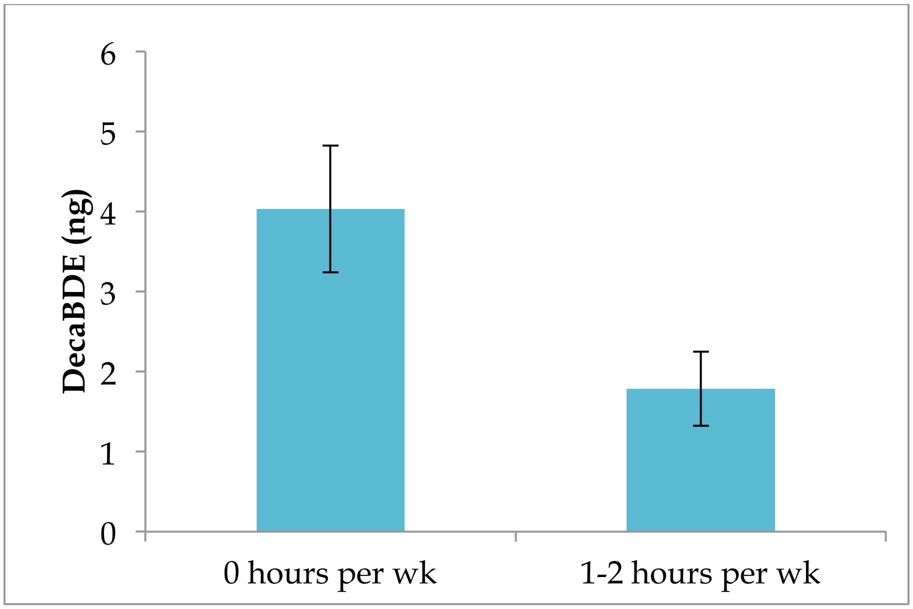

Figure 7. Comparison between DecaBDE levels on handwipes in individuals who reported spending zero, or 1-2 hours per week in a car.

Environment

The two variables examined for environment were time spent both indoors and outdoors. The variable for time spent indoors was split into 'low' (5-10 hours per day) and 'high' (>10 hours per day) and for outdoors into 'low' (<1 hour per day) and 'high' (1-5 hours per day). The low and high indoor group represented 32% and 68% of the population, respectively. The low and high outdoor group represented 12% and 88% of the population, respectively. The p-value for the differences between the outdoor groups was suggestive of relationship for PentaBDE (p-value=0.09) and the mean values were different by 40% (Figure 8). These data are consistent with other studies that point to the indoor environment as being important for exposure to PBDEs. Indeed, it seems the less time you spent outdoors (i.e. more time indoors), the higher the levels of exposure. Additionally, the differences between the indoor groups for both TBB and TBPH were not significant but highly correlated (p-value=0.14, p=0.12) with mean differences of 58 and 80%. These data may hint to and underlying relationship between time spent indoors and exposure to Firemaster® 550 or TBPH. More data is need, however, to investigate this potential relationship.

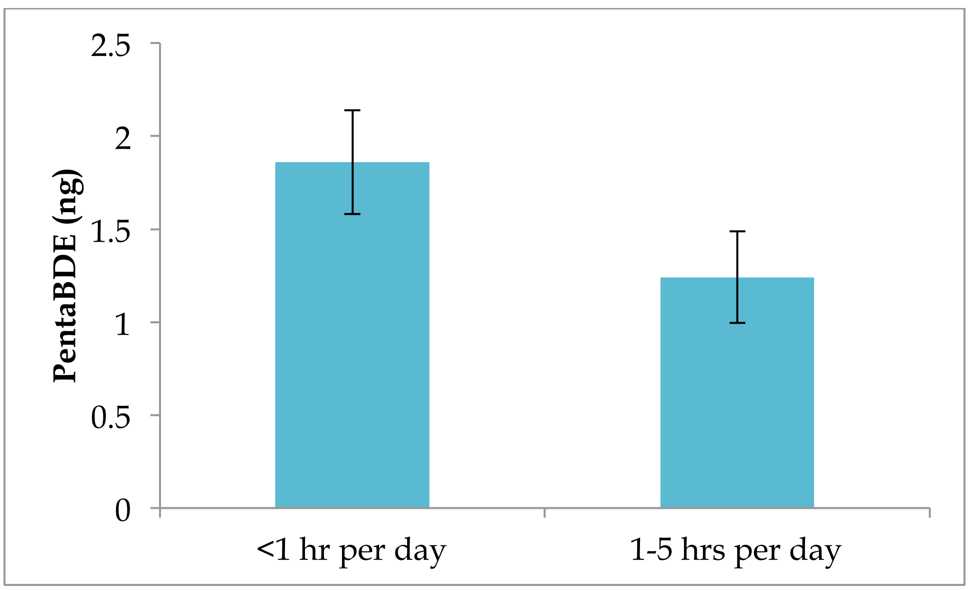

Figure 8. Comparison between PentaBDE levels in handwipes from individuals reporting different amounts of time spent outdoors.

Electronics

Several variables related to electronic item usage and ownership were examined including number of computers of TVs owned, time spent using the computer or TV, as well as the age of the computers owned. The majority of the sample population owned 1 or less computers (84%) and some owned more than one computer (16%). The difference in DecaBDE handwipe levels between the two groups was statistically significant (p-value=0.05) with a 77% difference in the means between the groups (Figure 9). For TBPH, the difference between the two groups was 78% but was not significant (p= 0.17). Time spent on the computer, whether ≤5 hours or >5 hours per day, was not significantly different between the two groups for any of the flame-retardants. Similarly, the age of the computer was not a significant predictor for any of the flame-retardants.

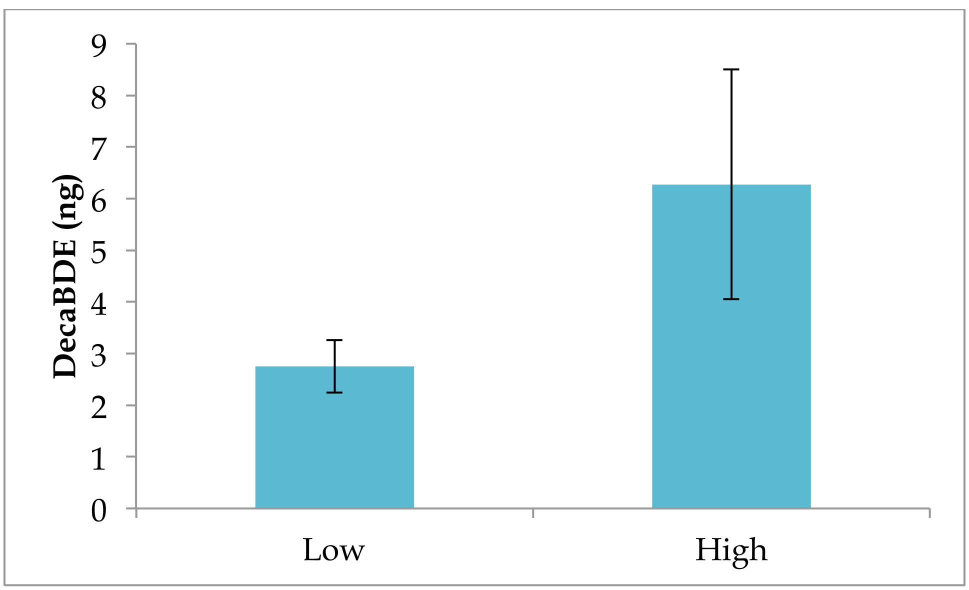

Figure 9. Comparison between DecaBDE levels on handwipes for individuals owning a high number of computers (>1), or low number of computers.

A majority of the sample population did not own a television (68%) and some owned one television (32%). Additionally, most students did not spend any time watching TV (64%) and only some spent time watching TV per day (36%). Television ownership was not a significant predictor of exposure to flame-retardant chemicals, perhaps because so few of the students actually owned a TV. Despite this, the p-values for DecaBDE were much lower between the groups than for Firemaster550 or TBPH. Furthermore, the levels of DecaBDE between the groups in the 'low' tv watching group versus the 'high' was suggestive of a relationship (p-value=0.11) (Figure 10). Moreover, the p-values for Firemaster550 and TBPH were very high compared to DecaBDE.

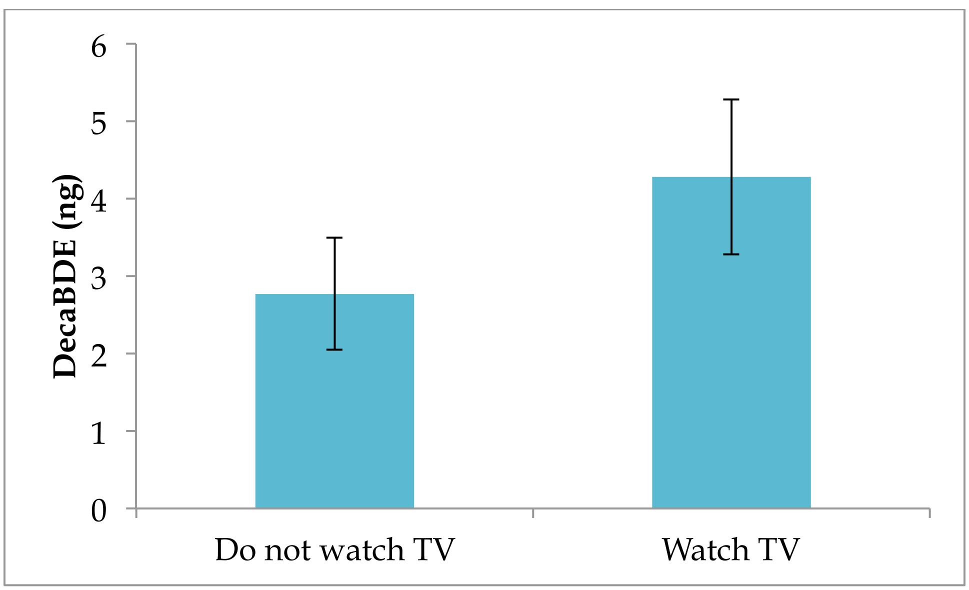

Figure 10. Comparison between DecaBDE handwipe levels in individuals that did or did not watch TV on a daily basis.

Examining these data, it appears that computer ownership and time spent watching TV are potentially important predictors of DecaBDE exposure even though only one of these relationships were significant. This is consistent with the usage patterns in China, especially for DecaBDE usage in electronic items. Furthermore, the data suggests that television ownership or usage is not a useful predictor of either Firemaster550 or TBPH concentrations. These compounds may not be used in these electronic items in China, but more research is needed to confirm this hypothesis.

Comparison to Other Studies

There are several studies that have measured PBDE levels in handwipe samples, however, they are limited to the United States (Stapleton, et al. 2012, Watkins, et al. 2011). The levels of PentaBDE are much lower from this study compared to levels found in the United States. Once again, this confirms the data that shows a higher usage of PentaBDE in the North America as compared to Asia. Furthermore, the levels of DecaBDE appear to be consistent between the two countries, confirming the similar usage patterns between the two regions. For TBB and TBPH, the levels between China and the U.S. are also consistent. It is still unknown, however, the exact production in these two countries and the products that may contain these compound. To date, there have been no studies measuring PBDE levels in handwipes from Chinese populations other than the present study.

Table 7. Comparison of PBDE values measured in handwipes from other studies

Source	PentaBDE		DecaBDE	Firemaster® 550	
	BDE 47	BDE 99	BDE 209	TBB	TBPH
Present Study (China)	0.26	0.16	2.1	0.35	2.3
Stapleton, et al. 2012 (United States)	16.3	13.8	2.0	4.2	2.4
Watkins, et al. 2011 (United States)	32.8	27.4	11.8	N/A	N/A

CONCLUSION

The small student population sampled from Shantou University in this study is exposed to both PBDEs and new use flame-retardants, TBB and TBPH. As expected, the levels of PentaBDE found in the handwipes from China were lower than in handwipes from similar studies performed in the United States. This is consistent with the fact that historically the North America, especially the U.S. has produced/used more PentaBDE than Asia. Levels for DecaBDE in handwipes were consistent with levels measured in the United States. This was expected due to similar usage behavior between the North Americas and Asia. Cleaning behaviors were the most significant predictors of exposure to PBDEs but not for compounds in Firemaster® 550. The more cleaning methods employed, the lower the exposure to PBDEs, likely due to the reduction of dust in the indoor environment and thus a reduction in exposure. Dust, however, may not be an important source of exposure to TBB and TBPH but more information should to be collected to confirm this. Behavioral variables such as time spent using electronic items and electronic item ownership generally are associated with higher DecaBDE levels but do not seem to predict levels of TBB and TBPH. The associated between electronic ownership and usage and the DecaBDE mixture is likely to it utilization primarily in the plastic casings of electronic items. The more interaction a person has with these items containing PBDEs, the greater the exposure to these chemicals. The reason for the lack of correlation to TBB and TBPH is unclear; it may be due to a difference in chemical properties of if these compounds or that they simply are not used in electronic goods in China. As replacements for PentaBDE, it is likely they are being used as flame-retardants in goods such textiles or furniture. Unlike other studies, hand washing was not a significant predictor of exposure. This, however, is most likely due to the high level of hand washing frequency among the study participants.

In the future, the levels found in the hand wipes from Shantou will be compared to the levels measured from the indoor dust from each participant. Furthermore, the levels of PBDEs measured from the student population from Shantou will be compared to levels measured from a student population from City University in Hong Kong. With a larger sample size, preliminary relationships between behavioral variables and levels of PBDEs may potentially be strengthened. Additionally, by comparing the two groups to each other, differences between the two urban areas and between the student populations may explain differences in exposure levels.

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APPENDIX

Consent Form

Primary Investigators: Lauren Gloekler, leg19@duke.edu; Margaret B. Murphy, mbmurphy.cityu@gmail.com; Ball Ching, ballching1973@yahoo.com.hk

Thank you for your interest in our project. You are being asked to participate in a study to determine exposure to flame retardant chemicals commonly found in indoor dust. This research study will collect 25 samples from City University of Hong Kong and 25 samples from Shantou University. Please read this consent form carefully and take your time making your decision. Please ask any member of the research staff to explain any words or information that you do not clearly understand.

GENERAL INFORMATION
This study will provide more knowledge about students' exposure to flame-retardant chemicals. If you agree to participate, we will collect a hand wipe sample from you today and collect a dust sample from your home and workplace. To collect a hand wipe sample, we will take a sterile gauze pad soaked in rubbing alcohol and wipe your hands. Collection of the hand wipe sample is painless and harmless. We will also collect dust from inside your home and workplace using our vacuum cleaner.

In addition, we will ask you to complete a survey about your activities and behaviors, as well as your home environment. The survey takes approximately 10 minutes to complete. The collection of hand wipe and dust samples will take approximately 20-30 minutes. We anticipate the whole process will take approximately 30-40 minutes.

Participation in this study is completely voluntary. You do not have to be in the study if you do not want to. If you agree to be in the study, you can skip any survey questions that you do not want to answer or you may withdraw from the study altogether by notifying one of the primary investigators, Lauren, Margaret, or Ball, at any time.

CONFIDENTIALITY
The information collected from you will be password-protected and will be maintained by the researchers participating in the study. Your name will be replaced by a unique code and will never be written on any sample we collect. We will report all data in an aggregated, de-identified way (i.e. not on an individual level). The study is a collaboration between City University and Shantou University, and therefore we may be asked to disclose the data we collect to participating staff at either university. If we do, we will only provide coded study data, and will never provide individual names or contact details.

We will keep your contact information in case we need to contact you in the future. We keep your contact information separate from your research data and in a password-protected file accessible only to the researchers in this study.

BENFITS AND RISKS/DISCOMFORTS
There are no direct benefits to you for participating in this study. However, the information we collect may help us determine exposure to flame retardant chemicals. We do not anticipate any risks or discomforts to you for choosing to be in this study.

***IMPORTANT NOTE:** It is unknown whether flame-retardants cause problems with human health.

FOLLOW UP
In approximately 9-12 months, you will receive information through email regarding the levels of flame-retardants measured in the wipe collected from your hands and what was measured in your dust. We will also let you know how the levels in your dust compare to levels measured in other homes and workplaces in the United States and provide you with contact information for follow-up should you have any questions.

CONTACT INFORMATION
For any questions about the study or if you have complaints, concerns or suggestions about the study, please contact Dr. Margaret Murphy at (852) 3442-6858 or Ball Ching at (852) 6833-2163 during regular business hours.

If you agree to be in the research, please check each below:

______ YES, I agree to participate in this research.

______ YES, I agree to allow the City University/ Shantou University research team to collect a hand wipe sample from me, as part of this research.

______ YES, I agree to allow the City University/ Shantou University research team to collect a dust sample from my home and workplace, as part of this research.

______ YES, I agree to allow the City University/ Shantou University to keep my contact information to re-contact me.

____ ____________

Signature of Student Date

Questionnaire for Examining Exposure to Novel Flame-Retardants

(Student Version)

Date:__________________ Recorded By:________________________

I. BACKGROUND INFORMATION

Full Name____ Height ___________ (cm)

Street Address_______________________________________ Weight __________ (kg)

City:_______________________ Province_______________ (Country)_________

Born in: (City, Province)______________

Age Group (Please select one):
______ < 18
______ 18-22
______ 23-26
______ 27-30
______ > 30

Marital Status: ____ Single _____Married ____Divorced _____ Widowed

Education (check all that apply):
____ Primary School
_____ Some High School
_____High School
_____ Some College
_____ Associates Degree
_____ Bachelor's Degree
_____ Graduate Deg. (e.g. Master's, Ph.D., M.D.)

II. WORK HISTORY

Employment: _______Employed ______ Unemployed

Do you work: _______ On Campus _______ Off Campus

Where do you work? ________________________________

How many hours per week do you work? ________________

Do you have any other jobs? If so, please describe and state how many hours per week you work in

this position. ____

III. DIET

General

On average, how many of the meals you eat every week are cooked at home?

_____ None (0) ______ Some (1-5) _______ Many (5-10) ______ Almost All (>10)

On average, how many of the meals you eat per week are at a restaurant/food court/or street food vender?

_____ None (0) ______ Some (1-5) _______ Many (5-10) ______ Almost All (>10)

On average, how many meals do you eat per week at a fast food restaurant (KFC, McDonald's, Burger King)?

_____ None (0) ______ Some (1-5) _______ Many (5-10) ______ Almost All (>10)

Meat/Seafood

On average, how many servings (85 grams per serving) of the following meat/meat products do you consume per week?

Meat Type	**Never (0 servings)**	**Rarely (1-2 servings)**	**Occasionally (3-5 servings)**	**Frequently (>5 servings)**
Beef (*e.g. meatball, sliced, rib*)				
Chicken (*e.g. wing, leg, sliced, feet*)				
Pork (*e.g. chop, sausage, rib, bacon*)				
Duck (*e.g.*				

breast, leg)				
Quail (*e.g. whole, leg*)				
Lamb (*e.g. leg, loin, chop, rib*)				
Eggs (*e.g. Chicken, Quail, Duck, etc*)				
Organs (*e.g. stomach, intestine, liver, kidney*)				
Other:				
Other:				

Fish/Seafood

On average, how many servings (85 grams per serving) of the following shellfish/seafood items do you eat per week?

Type of Seafood	**Never (0 servings)**	**Rarely (1-2 servings)**	**Occasionally (3-5 servings)**	**Frequently (>5 servings)**
Clam				
Crab				
Cuttlefish				
Eel				
Fish				
Jellyfish				
Lobster				
Octopus				
Mussel				

Sea Snail				
Squid				
Scallop				
Shrimp				
Other:				
Other:				

Dairy

On average, how many servings of the following dairy foods (227 grams per serving) do you consume per week?

Type of Dairy	**Never (0 servings)**	**Rarely (1-2 servings)**	**Occasionally (3-5 servings)**	**Frequently (>5 servings)**
Whole Milk				
Skim Milk				
Condensed Milk				
Ice Cream				
Yogurt				
Cheese				
Other:				

Rice/Noodle

On average, how many servings (114 grams per serving) of the following items do you eat per week?

Type of Food	**Never (0 servings)**	**Rarely (1-2 servings)**	**Occasionally (3-5 servings)**	**Frequently (>5 servings)**
Rice				
Congee				
Noodle				

Beverages

How many servings (227 grams per serving) of each beverage type do you typically consume per week?

Type	**Never (0 servings)**	**Rarely (1-2 servings)**	**Occasionally (3-5 servings)**	**Frequently (>5 servings)**
Water				
Tea				
Coffee				
Soda (Coke, Sprite, etc)				
Fruit Juice				
Alcoholic (beer, wine, liquor)				

IV. Behavioral Traits

Do you currently smoke? _____ Yes _____ No

Have you smoked in the past? _____ Yes _____ No
If yes, for how long? (State your answer in years or months)__________________________

How long ago did you quit smoking? (State your answer in years or months)_____________

Do you currently live with others who smoke? ____ Yes ____ No
If yes, how many people?___________

Do you bite your nails? ____ Yes ____ No ____ Sometimes

Do you chew your hair? ____ Yes ____ No ____ Sometimes

Do you commonly lick your fingers after eating finger foods? ____ Yes ____ No ____ Sometimes

How many times a day do you wash your hands?
_____None ____ 1-2 times/day ____ 3-4 times/day _____ > 4 times/day

How many hours a day do you spend outdoors?

_____Less than 1 hour _______ 1-5 hours _______ 5-10 hours ________ More than 10 hours

How many hours a day do you spend indoors?

_____Less than 1 hour _______ 1-5 hours _______ 5-10 hours ________ More than 10 hours

On average, how much time, in hours, do you spend using each form of transportation per week?

Transportation Type	**Never (0 hours/week)**	**Rarely (1-2 hours/week)**	**Occasionally (3-5 hours/week)**	**Frequently (>5 hours/week)**
Bus				
Ferry				
Car (Personal)				
Metro (MTR)				
Taxi				
Other:				

Do you have any pets? _____ Yes ____ No
If yes, how many do you have: ____ Dogs ____Cats ____ Other

V. HOME ENVIRONMENT

Dwelling

Do you live in a (check one):

______ Dorm ______ Apartment ______Government Housing ______ Townhouse ______House

______Other (Please describe)____

How long have you lived in your current residence? ____________________ years or months

Is your place of residence (check one):

_____ Mainly carpeted (e.g. more than 50%)
_____ Mainly wood floors (e.g. more than 50%)
_____ Mainly tiled (e.g. more than 50%)
_____ Mixed carpet/wood floors (equal wooded and carpeted floors)
_____ Mixed carpet/tiled floors (equal carpeted and tiled floors)
_____ Other (Please describe)___________________________________

Does your home have (check one):

_____ Central air conditioning
_____ Window air conditioning units (how many_______)
_____ No cooling system

How many times a week do you clean your home?
_____none ____ 1-2 times/week ____ 3-4 times/week _____ > 4 times/week

What do you typically use to clean the floors in your home? (If applicable, check more than one)

______Broom ______Vacuum _______ Mop/Swiffer ______ Other (Please describe)__________

What do you typically use to clean hard surfaces/furniture in your home?

______Dust spray _______Feather duster ______Soap & water ______Other (Please describe)____________

Electronic Goods/Furniture

How many of the following electronic items do you have? Please include those present in both your home and work environment.

Electronic Item	**None**	**One**	**Two**	**Other (Please specify)**
Television				
Computer				
iPad/ iPhone				
Telephone				
Printer/Copier				

Fax Machine				
Gaming System (Wii, Playstation, X-Box)				
Other:				

If you have a TV, how many hours per day is the TV on? __________ hours
Approximately how old is your television? _________ years
If you have a computer, how many hours per day are you using the computer (include use during work)________________________
What type of computer/s do you own (Mac/PC)? _____________
Approximately how old is your computer? ____________ years

Have you purchased any new furniture in the last year, such as new living room furniture, a new mattress, new carpets/floors, a new TV, ? _____ Yes _____ No _____ Don't know
If yes, please list what items are new____
____