Title:

Preliminary Examination of the socioeconomic and housing factors that influence exposure to flame-retardants (FRs) in passive samplers worn by preschool-aged children in Oregon

By

Kenneth Willard

An Undergraduate Thesis Submitted to

Oregon State University

In partial fulfillment of

the requirements for the

degree of

Baccalaureate of Science in BioResource Research,

Toxicology Option

10/31/14

Signature Page:

APPROVED:

___________________________________ ________________

Dr. Molly Kile, Public Health and Human Sciences Date

___________________________________ ________________

Dr. Kim Anderson, Environmental and Molecular Toxicology Date

___________________________________ ________________

Katharine G. Field, BRR Director Date

I understand that my project will become part of the permanent collection of the Oregon State University Library, and will become part of the Scholars Archive collection for BioResource Research. My signature below authorizes release of my project and thesis to any reader upon request.

__________________________________ ________________

Kenneth Willard Date

Table of Contents

Abstract

Flame-retardants (FRs) generally enter the environment by dissociating from the products they were originally manufactured into. The predominant class of FR used in consumer products before their eventual phase-out in the mid-2000s were the polybrominated diphenyl ethers (PBDEs). Previous studies have illustrated a correlation between socio-economic status, notably gross income, and exposures to PBDEs. This study examined five congeners of PBDEs as well as four congeners of the replacement FRs. To accomplish this, silicone passive sampling bracelets, developed at Oregon State by Dr. Kim Anderson, were worn by 83 pre-school aged children for up to one week. Once the bracelets were analyzed, we used the data to examine correlations between the loading rate of the nine congeners and nine socio-economic factors that we selected based on past studies. This study found a significant correlation between the amount of times a person vacuumed their homes and an increased loading rate of FRs, as well as a correlation with gross household income.

Keywords: Flame-retardants (FRs), Polybrominated diphenyl ethers (PBDEs), passive sampling

1. Introduction

1.1 Polybrominated Diphenyl Ethers (PBDEs)

Polybrominated diphenyl ethers (PBDEs) are classified as persistent organic pollutants under the Stockholm Convention. Consisting of two phenyl groups with a varying number of bromines attached, the compounds are highly hydrophobic and hydrophobicity increases with the amount of bromines that are bonded to them.[1] They are used as flame retardants in the manufacturing of electronics and in household furnishings and are a part of a class of compounds known as brominated flame retardants (BFRs). BFRs are added to products in the manufacturing process to try and slow the process of combustion. They achieve this goal by bonding the free radicals (generally oxygen molecules) and slowing the spread of the fire.[2] Bromines can be added into the compound in a variety of ways. The method used for producing PBDEs is referred to as the additive method and works by mixing in the PBDE compounds into the polymers. Reactive addition and brominated monomers are other common ways for these compounds to be added to products.[2]

PBDEs are classified by the number of bromine atoms they contain. The three most common groups found in consumer products are the penta-, octa-, and deca- mixtures which have five, eight and ten bromines, respectively. The different number of bromines attached to the base diphenyl-ether molecule can affect their toxicity. For example, in laboratory animal studies it has been demonstrated that penta-PBDEs are more likely to cause neurological developmental problems, while the more brominated deca-PBDEs have the potential for tumor neogenesis.[3] When PBDEs were added to a product, it is never just a single congener of PBDE. For instance, the commercial product, PentaPBDE (BK-70®), is made up of primarily two congeners, PBDE-47 and PBDE-99, which accounts for almost 70% of its mass by volume. The other two predominant congeners are hexa-PBDEs 153 and 154. PentaPBDE was the most widely used brominated flame retardant product in the United States[4] and we will focus on these four congeners in this study.

1.2 Exposure Routes of PBDEs

Products that contain polyurethane foams are of special concern because of the amount of PBDEs that are added into them in manufacturing.[5] PBDEs are not as tightly bound to the products they are in, physical agitation can be enough to make them dissociate from the product.[6] Once they enter into the environment, the primary route of exposure for humans is thought to be ingestion of house dust.[6] This is one reason why the United States Environmental Protection Agency recommends using a damp mop on hardwood floors and a vacuum with a HEPA quality filter to vacuum carpets to reduce exposure to PBDEs.[7]Additionally, another route of exposure for PBDEs is inhalation of house dust and vapors. Multiple studies have demonstrated that lower-income houses are at higher risk for PBDE exposures[89]. It has also been demonstrated that households with a higher percentage of exposed polyurethane foam have higher exposures to PBDEs[9].

Children are considered to be a susceptible sub-population in regards to PBDE exposure and its related toxicity. A study completed in 2003 examined the concentrations of PBDEs in the blood serum of pregnant women and their developing fetuses. The study indicated that maternal PBDE blood levels are good indicators of fetal levels and that PBDEs cross the placenta during pregnancy.[10] Since PBDEs are highly lipophilic they are also present in breastmilk.[11] These factors, in addition to the reduced body size of children and their propensity for age-specific behaviors such as more hand-to-mouth activity, and crawling, put infants and young children at a higher risk for PBDE exposure[12]. It has been shown that the levels of PBDEs being found in children can exceed the EPA recommended levels.[8] Children also have a smaller body mass than adults which would result in a higher dose to the child from a similar environmental concentration. Children also have under developed immune systems and detoxifying mechanisms.

1.3 Regulation of PBDEs

On February 6th 2003 the European Chemicals Agency (ECHA) proposed that the EU put heavy restrictions on the use of penta and octa PBDEs, banning the sale of any product that contained more the 0.1% of the mass of either chemical.[13] This legislation was brought upon after multiple studies demonstrated rising PBDE concentrations in humans since their use began in the 1970s. This was the first time regulation had been passed specifically targeting the sale and use of PBDEs. Following the lead of ECHA and the EU, in August of 2003, California legislatures signed a bill into law that banned penta- and octaPBDEs from being added into products. Due to a compromise between BFR manufacturers and lawmakers, the official ban would not be enforced until 2008. However, California's motion to limit the use of BFR's started a trend that would soon have many states reconsidering their position on the use of these products.

Closely after the California legislation was completed, the United States Environmental Protection Agency (EPA) drafted new legislation aimed to phase out the production and inclusion of

PentaPBDE and octaPBDE in consumer products by January 1st, 2005. The legislation required the major producers of brominated flame retardants to cease manufacturing these compounds, unless the reason for using PBDES qualified for a significant new use ruling. This allowed the EPA to closely examine any new manufacturing of these chemicals before they would hit the market. The manufacturer would also have to demonstrate that the chemical was needed and could not be replaced. The chemical companies agreed to the voluntary phase out of the octa- and penta-PBDE products by January 1st, 2005.

In 2013, California revised its Technical Bulletin 117 that was enacted in 1975 to remove the requirement for the addition of flame retardant chemicals to pieces of furniture. Coinciding with this revision, the EPA also updated its stance on PBDEs stating that any use of penta, octa, or decaPBDEs qualified as a SNUR and would be subject to EPA oversight in its manufacturing. The new plan also requires manufacturers to provide more information on the production and transportation of the chemicals.

1.4 Use of silicone passive samplers

This pilot study was interested in determining the presence of PBDEs in the air that would be relevant to children. To accomplish this our study used silicone based passive samplers which were worn around the wrist or ankle of the children for one week. The passive sampling devices (PSDs) were designed at Oregon State University by Dr. Kim Anderson. The bands were capable of sequestering 41 flame-retardant compounds and showed spatial and temporal sensitivity which allowed for a more rigorous assessment of the child's microenvironment[14].

1.5 Focus of this study

This study will focus on a selection of socioeconomic and housing factors that may influence children's exposure to PBDEs in the air. We used passive samplers worn by preschool aged children that sample volatile PBDEs to measure personal exposure to these compounds. . The socioeconomic and housing factors we have chosen were selected because we thought they might have the greatest impact on the loading rates of PBDEs to the passive sampling devices. The selected factors were chosen based on a questionnaire that was given to the parents of the children prior to sampling. The questions were asked to gain a broader understanding of the environment that the children were experiencing on a day to day basis. Specifically, we asked participants how many times a month they vacuumed and mopped their homes because of the EPA's recommendation that these behaviors could reduce exposure to PBDEs[7]. The age and size of the home was also considered as potential factors that could influence PBDE exposure since PBDE mixtures have faced different restrictions over time. This was based on the work by Zota et al. and their work detailing the decline in human concentrations in the years following the ban of PBDEs by California and the EPA.[15] The type of flooring that was in the house, particularly how much carpet was in the house, was of particular interest because previous studies have shown that carpet was a reservoir for PBDEs[16] The amount of carpet is especially important when looking at the exposures of children because of their increased time spent near the ground and the increased hand to mouth activity. The type of pillow and mattress that the child was looked at because of amount of polyurethane foam that is contained in them and their close proximity to the breathing zone during sleep[17] Finally, gross household income was examined because previous research has identified income as a possible correlating factor in PBDE body burden.[18]

1.6 Hypotheses

The hypotheses examined in this thesis include:

- Children that live in homes that are vacuumed more frequently are anticipated to have a lower loading rate of flame-retardants onto the wristbands, per the EPA guidelines for reducing exposures[7], compared to children that live in homes that are vacuumed less frequently.
- Children that live in homes that are mopped more frequently are anticipated to have lower loading rates of flame-retardants onto the wristbands compared to children that live in homes

that are mopped less frequently.

- Children that lived in a house that was built between 1976-2005 are anticipated to have a higher loading rate of flame-retardants onto the wristbands because of the timing of the legislation of flame-retardants by the Federal and state governments compared to those houses were built before 1976 and after 2005.
- Children that live in smaller homes are anticipated to have higher loading rates of flame-retardants compared to children who live in larger homes.
- Children that live in homes that have a higher percentage of the floor covered in carpet are anticipated to have a higher loading rate of flame-retardants because of the excess polyurethane foam that is present in carpeting compared to houses that have a higher percentage of wood or vinyl floorings.
- Children who live in households with lower gross income are anticipated to have a higher loading rate of flame-retardants compared to children who live in households with higher gross income.

2. Materials and Methods

2.1 Population characteristics

Children were from two communities - Bend and Corvallis, Oregon. In total we had 28 participants in Corvallis and 65 in Bend. Children were recruited in their first year of preschool and were aged 3 to 5 years. This was not a random sample.

2.2 Use of Silicone Passive Samplers

This pilot study was interested in determining the amount of PBDEs that a child came in contact with in the air over the course of seven days.. Each child was given a wristband at the end of the home visit. Parents and the child were briefed on the what the band was, how it could be worn, and what it would be sampling. Each child was instructed to wear the band for 7 days on either the wrist or ankle. Participants could wear the band while doing any activity but were also told they could take the band off when bathing or sleeping, as long as the band was in the environment of the child as much as possible. Parents were asked to record the number of days that the child wore the bracelet on the self-enclosed label and enclose it in the provided PTFE airtight bag after the child worn the band. As a result of the voluntary participation, we had an attrition rate of 14% of the bands not being returned, making the final testable population N=77.

The passive sampling devices (PSDs) were designed at Oregon State University by Dr. Kim Anderson. The bands were capable of sequestering 41 flame-retardant compounds and showed spatial and temporal variability which allowed for a more rigorous assessment of the child's microenvironment[14].

2.3 Chemical Analysis

The parents were given an air-tight bag and told to seal the wristbands in the bag after 7 days. These bags were then then placed in a provided, self-addressed envelope and mailed to the project office at Oregon State University. The bands were logged in when they were received and the number of days the child wore the band were recorded. The wristbands were then transferred to Dr Kim Anderson's lab following a chain-of-custody protocol for analysis. All chemical quantitative analysis was conducted via processes developed by Dr. Anderson's Laboratory. The following link provides more detail on Dr. Anderson's laboratory at Oregon State University: http://fses.oregonstate.edu/Kim-Anderson Details of the chemical extraction will be the subject of a future publication by Dr Anderson.

2.4 Questionnaire

Parents were given a questionnaire at the initial home visit. The questionnaire was designed to give insight on the subjects' home life and living conditions. From this questionnaire, nine questions were selected to be researched. Those questions were:

- Participants were able to choose between four options on the questionnaire for mattress type: Foam, Spring, Mix and Unsure.
- Participants had to choose one of five options in pillow type: Foam, Synthetic, Down, Mix and Unsure.
- The responses for both vacuuming and mopping were able to be self-reported on the questionnaire.
- Participants were asked to list all of the types of flooring that was in their home
- Participants were asked to estimate the percentage of their total floor space that was covered by carpet.
- Participants were asked to self-report the age of their home to the nearest year.
- Participants were asked to self-report how big their home was in square feet.
- When examining income levels we gave participants to choose between 8 categories: less than $22,000, $22,001-$30,000, $30,001-38,000, $38,001-46,000 , $46,001-54,000 , $54,001-62,000 , $62,001-70,000 and $70,000+.

2.5 Statistical analysis

Data from the 77 wristbands and survey answers was imported into IBM SPSS statistical software for easier analysis. A two-tailed T-test was used to determine if there was any statistical difference between the two communities. Once it was determined that there was no significant difference in the two geographical areas (TCEP $t(77)=.31$, $p=.68$; TCPP $t(77)=.68$, $p=.52$; TDCPP $t(77)=.72$., p=.31; TPP t(77)=., p=.90; PBDE 47 t(77)=.75, p=.17; PBDE100 t(77)=.55, p=.10; PBDE99 t(77)=.4, p=.21; PBDE 154 t(77)=.63, p=.78; PBDE 153 t(77)=.64, p=.77), we combined the groups to allow for statistical power in our tests. For comparisons that had more than two groups, we used analysis of variance (ANOVA) .Since many of the selected factors had few samples in a specific group, we collapsed data into categories to maximize sample size as follows:

2.5.1 Mattress type

Only three participants indicated that they slept on a mixture of mattress types. To improve the power of the statistical test, we combined the group of mix and unsure to create a dummy statistic "Mix+Unsure".

2.5.2 Pillow type

The decision was made to merge foam and synthetic into one category: "Foam+Synthetic". This was for two reasons; only two people indicated that they slept on a foam pillow which would have negatively impacted the confidence in the statistical test and secondly because the materials that fill both synthetic and foam pillows are very similar (polyurethane foams). We decided against merging the down pillow category with another even though the sample size was low (n=3) because of the uniqueness of the down pillow filling.

2.5.3 Vacuuming and Mopping.

The responses for both vacuuming and mopping were able to be self-reported on the questionnaire. Because of the multitude of unique we decided to report the answers as an average amount of times mopping or vacuuming per week. The answers were placed in three categories for each : 1-4 times per month (average of once a week), 5-8 times per month (average of two times a week), and 9+ times per month (3+ times per week).

In the case of mopping, fewer people answered that they mopped their homes more than three times per week so to facilitate the statistical tests we decided to run a two-tailed T-test comparing the groups: 1-4 times per month, and more than 5 times per month.

2.5.4 Income level

When examining income levels we gave participants to choose between 8 categories. In looking at the raw numbers we realized that we had multiple categories with low sample sizes ($n<5$). We merged the middle 6 categories into one category: "$22,001-$70,000" to provide us greater sample size ($n=25$). This gave us a total of three categories with greater numbers in the sample and still allowed us to observe the effects of income on the loading rates of flame-retardants.

All other categories that were analyzed were kept as they were reported by the participants. No other categories were merged in our final analysis.

3. Results and Discussion

The purpose of this study was to determine whether common housing and socio-economic characteristics were associated with commonly used flame retardants in the air. The five PBDE congeners (PBDE 47, PBDE 99, PBDE 100, PBDE 153, PBDE 154) were selected based on their reported use in industry, namely in the product BK-70® which is colloquially known as PentaPBDE. Also analyzed were the "tris" compounds (TCEP, TPP, TCPP, TDCPP) that have been used to replace the PBDEs as flame retardants.

3.1 Analysis of Pillows and Mattress types

The first analysis evaluated whether the type of mattress that a child slept was associated with average daily FR concentrations. The responses were grouped into three mattress types: Foam, spring and unsure (refer to Table 1 for population information). Participants that marked both spring and foam were placed in a fourth category called "mixed". The average daily FR concentrations measured in the bracelet did not differ between foam, spring, or unsure mattress types, TCEP $F(2, 74)=.58$, $p=.56$; TCPP

F(2,74)=.66, p=.52; TDCPP F(2,74)=.61, p=.55; TPP F(2,74)=.20, p=.82; PBDE 47 F(2,74)=.23, p=.80; PBDE100 F(2,74)=.25, p=.78; PBDE99 F(2,74)=.11, p=.89; PBDE 154 F(2,74)=.55, p=.58; PBDE 153 F(2,74)=1.1, p=.33. (Fig. 2)

In the same vein as mattress types, we attempted to determine whether the average daily FR concentration measured in the bracelets was associated with the type of pillow a child used. The results show that there is no significant relationship between pillow type and the average daily congener concentrations measured in the bracelets, TCEP F(3,73)=2.32, p=.08; TCPP F(3,73)=2.201, p=.095; TDCPP F(3,73)=.43, p=.73; TPP F(3,73)=1.33, p=.27; PBDE 47 F(3,73)=1.63, p=.19; PBDE100 F(3,73)=1.63, p=.19; PBDE99 F(3,73)=1.42, p=.24; PBDE 154 F(3,73)=1.26, p=.29; PBDE 153 F(3,73)=1.40, p=.25. (Fig. 3)

These analyses suggested that average daily concentration measured in the child's breathing zone did not differ based on the mattress type or the pillow type. Although the subsequent statistical tests revealed no significant differences between the different materials, this could be due to the common practice of covering pillows and mattresses. It would be interesting to measure the concentration of flame retardants in the actual products to see if the constituency of the material differed in these products. Multiple contaminants were approaching significance, however the power of the statistical test was reduced because of small sample sizes. While none of the results returned significant differences, mattress and pillow materials would be a good area of future research since our study suffered from a lack of variability in the types of mattresses and pillows used with a majority of the participants indicating that they slept on a mixture of pillow and mattress types. With a larger sample of people who sleep on exclusively foam or spring mattresses, it may yield a different results. This is true with pillows as well. Because of the amount of time kids, and people in general, spend sleeping it would be wise for future research to explore the effects of the materials we sleep on with regards to our chemical burdens.

3.2 Analysis of Floor Type and Carpet Coverage

The type of floor that is in a home can have an impact on the indoor environment of the home. The majority of the people in the survey answered that they had a mix of floorings in their home (n=66), while only 10 subjects had a singular flooring in their home (carpet, n=5; hardwood, n=5). A comparison of these three groups (mixed, carpet and hardwood) showed that there was no significant difference in the mean concentrations of each compound between the groups, TCEP $F(2,73)=.61$, $p=.55$; TCPP $F(2,73)=1.48$, $p=.23$; TDCPP $F(2,73)=.42$, $p=.66$; TPP $F(2,73)=.80$, $p=.45$; PBDE 47 $F(2,73)=1.93$, $p=.15$; PBDE100 $F(2,73)=2.23$, $p=.12$; PBDE99 $F(2,73)=1.85$, $p=.16$; PBDE 154 $F(2,73)=1.10$, $p=.34$; PBDE 153 $F(2,73)=1.18$, $p=.31$. (Fig. 4)

To determine if carpet has any distinguishable effect on the loading rates of FRs, we asked participants to indicate how much of their home is carpeted. Carpet has both fibers in its top layer and foam padding used to insulate it. It was thought that carpet may be a good reservoir for flame retardants. As stated earlier, a majority of the participants in this study indicated that they had a mixture of flooring types with 84% of them stating they had at least some carpet in their home. The mean concentrations of each congeners did not differ significantly based on the percentage of carpet covering the floor, TCEP $F(3,73)=.41$, $p=.75$; TCPP $F(3,73)=1.43$, $p=.24$; TDCPP $F(3,73)=.39$, $p=.76$; TPP $F(3,73)=.04$, $p=.99$; PBDE 47 $F(3,73)=.06$, $p=.98$; PBDE100 $F(3,73)=.12$, $p=.95$; PBDE99 $F(3,73)=.05$, $p=.99$; PBDE 154 $F(3,73)=.24$, $p=.87$; PBDE 153 $F(3,73)=.48$, $p=.70$. (Fig. 5) Future studies should analyze house dust to further explore possible difference in the various flooring types seen in homes.

3.3 Analysis of Size and Age of Home

This test was done to see if the size of the house had any effect on the loading rate of the FRs onto the bands. Participants were delineated by the size of their house, measured by square footage estimates done by the participants themselves. There were no statistically significant differences in mean level of each congener between the five groups of house size, TCEP $F(4,64)=1.08$, $p=.38$; TCPP $F(4,64)=.37$, $p=.83$; TDCPP $F(4,64)=1.02$, $p=.40$; TPP $F(4,64)=2.06$, $p=.10$; PBDE 47 $F(4,64)=1.50$, $p=.21$; PBDE100 $F(4,64)=1.04$, $p=.40$; PBDE99 $F(4,64)=1.59$, $p=.19$; PBDE 154 $F(4,64)=1.45$, $p=.28$; PBDE 153 $F(4,64)=.70$, $p=.60$. (Fig. 6)

We asked participants to indicate when their home was built in order to analyze accumulation over time. There were no statistically significant differences in mean level of each congener between the three groups of home age, TCEP $F(2,50)=1.54$, $p=.22$; TCPP $F(2,50)=1.24$, $p=.30$; TDCPP $F(2,50)=2.18$, $p=.12$; TPP $F(2,50)=.16$, $p=.86$; PBDE 47 $F(2,50)=.78$, $p=.46$; PBDE100 $F(2,50)=.91$, $p=.41$; PBDE99 $F(2,50)=1.61$, $p=.21$; PBDE 154 $F(2,50)=.62$, $p=.54$; PBDE 153 $F(2,50)=.33$, $p=.72$. (Fig. 7)

3.4 Analysis of Mopping and Vacuuming

Participants were asked to estimate how many times a month they vacuumed their homes. The data show that those who vacuum their house the most often have, on average, higher levels of PBDE 153 and PBDE 154 loading on to the bands, TCEP $F(2,74)=1.07$, $p=.35$; TCPP $F(2,74)=2.27$, $p=.11$; TDCPP $F(2,74)=.26$, $p=.78$; TPP $F(2,74)=.78$, $p=.46$; PBDE 47 $F(2,74)=2.30$, $p=.10$; PBDE100 $F(2,74)=1.28$, $p=.28$; PBDE99 $F(2,74)=2.83$, $p=.07$; PBDE 154 $F(2,74)=3.76$, $p=.03$; PBDE 153 $F(2,74)=4.56$, $p=.01$. (Fig. 8) Compared to participants that reported that reported vacuuming their home 0-4 times/month. Additionally, there appears to be a slight, albeit non-significant trend of higher levels of chemical loading with increased vacuuming appears for all contaminants except for TCPP.

One possible explanation for this finding is that the passive samplers sequester semi-volatile compounds. Inadequate vacuuming could lead to an increase of the dust content in the indoor atmosphere of the home and subsequently a greater reservoir of dust of volatilization. It is also possible that more frequent vacuuming volatilizes ultra-fine particles that are not trapped by the vacuum which directly increases the concentration of these compounds in indoor air which is then detected by the passive sampling devices. Future research should compare the collected house dust samples and the loading rates on the passive samplers to see if the opposite relationship is present.

We also examined the amount of times participants mopped their homes a month. The average congener level did not significant differ between the two groups who mopped on average once a week or more than once a week, TCEP t(73)=.31, p=.58; TCPP t(73)=.68, p=.41; TDCPP t(73)=.72., p=.4; TPP t(73)=1.36, p=.25; PBDE 47 t(73)=.75, p=.39; PBDE100 t(73)=.55, p=.46; PBDE99 t(73)=.00, p=.97; PBDE 154 t(73)=.03, p=.86; PBDE 153 t(73)=.04, p=.85. (Fig. 9)

There are a few possible explanations for this divergence between mopping and vacuuming, even though both are common house cleaning procedures. Mopping does not blow air, whereas, vacuuming may disperse particles into the indoor atmosphere lead to the increase in flame retardant loading rates among those who vacuumed more often. With mopping, there is no mixing of the indoor air occurring which could explain the relatively little effect mopping has on the loading rates of the flame retardants.

3.5 Analysis of Household Income

This analysis observed that people who are in the lower economic tiers tend to have a higher average contaminant loading in the bands than individuals who had a higher household income, TCEP F(2,74)=.04, p=.06; TCPP F(2,74)=5.76, p=.01; TDCPP F(2,74)=3.20, p=.05; TPP F(2,74)=5.32, p=.01; PBDE 47 F(2,74)=4.02, p=.02; PBDE100 F(2,74)=3.6, p=.03; PBDE99 F(2,74)=3.46, p=.04; PBDE 154 F(2,74)=7.02, p=.00; PBDE 153 F(2,74)=6.38, p=.00. (Fig. 10). It is also interesting to note that when looking at only the chlorinated flame retardants, the magnitude of the difference in the means becomes much smaller than when comparing the PBDE congeners across the same groups.

The data paints a slightly different picture when looking at the “tris” compounds. The “tris” compounds, which are supposed to be replacing the brominated flame retardants, were found in similar magnitudes as the brominated compounds. Every compound except for TCEP was found to have a statistically significant difference in their means between groups. However, the spread of the means between the groups is greatly reduced. This could be due to a number of factors. It is possible that with the less affluent participants, many of them are buying furniture and living in homes that are generally older. These items, which were made to meet TB117, could contain a higher concentration of PBDEs. Newer products, those manufactured after the repeal of TB117, would be more likely to have more of the chlorinated compounds in them, than the PBDEs. With a higher income, it would be expected that items would be bought new, and might have a higher turnover rate (replacing furniture more rapidly, etc.) which would lead to the higher amounts of chlorinated flame-retardants in their homes. This would also account for why the higher income families tended to have a lower rate of PBDEs loading onto the bands.

Based on the results of this study, socio-economic status seems to be a factor in children's exposures to flame-retardants, and consequently the loading rate of the flame-retardants onto the bands. Future research should look to gather participants from a larger geographic area since all subjects were recruited from western Oregon and may not be representative of a larger population.

4. Conclusion

This pilot study provides a brief insight into housing and socioeconomic characteristics that could influence children's exposure to polybromindated diphenyl ethers and chlorinated flame retardants in the air. Our study found that there is a relationship between the income of the household and the loading rates of the FRs onto the wristbands. Our study also illustrated an increased loading rate associated with an increase in the amount of times a family vacuumed their house per month. Further research should be done to determine effective measures of reducing FR exposures in the home.

We hope that the information gathered will be useful in conjunction with house-dust samples that were taken congruently to the passive sampling events. This information will be used to explore a possible relationship between FR exposures and learning deficiencies in young children.

5. References

(1) Gouin, T.; Harner, T. Modelling the environmental fate of the polybrominated diphenyl ethers. *Environ. Int.* **2003**, *29*, 717–724.

(2) Alaee, M.; Arias, P.; Sjödin, A.; Bergman, Å. An overview of commercially used brominated flame retardants, their applications, their use patterns in different countries/regions and possible modes of release. *Environ. Int.* **2003**, *29*, 683–689.

(3) ATSDR - Public Health Statement: Polybrominated Biphenyls (PBBs) & Polybrominated Diphenyl Ethers (PBDEs) http://www.atsdr.cdc.gov/phs/phs.asp?id=899&tid=94 (accessed Jan 29, 2014).

(4) Birnbaum, L. S.; Staskal, D. F. Brominated flame retardants: cause for concern? *Environ. Health Perspect.* **2004**, *112*, 9–17.

(5) Hallgren, S.; Darnerud, P. O. Polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs) and chlorinated paraffins (CPs) in rats—testing interactions and mechanisms for thyroid hormone effects. *Toxicology* **2002**, *177*, 227–243.

(6) Mazdai, A.; Dodder, N. G.; Abernathy, M. P.; Hites, R. A.; Bigsby, R. M. Polybrominated diphenyl ethers in maternal and fetal blood samples. *Environ. Health Perspect.* **2003**, *111*, 1249–1252.

(7) Schecter, A.; Päpke, O.; Tung, K.-C.; Staskal, D.; Birnbaum, L. Polybrominated Diphenyl Ethers Contamination of United States Food. *Environ. Sci. Technol.* **2004**, *38*, 5306–5311.

(8) Stapleton, H. M.; Sharma, S.; Getzinger, G.; Ferguson, P. L.; Gabriel, M.; Webster, T. F.; Blum, A. Novel and High Volume Use Flame Retardants in US Couches Reflective of the 2005 PentaBDE Phase Out. *Environ. Sci. Technol.* **2012**, *46*, 13432–13439.

(9) Jones-Otazo, H. A.; Clarke, J. P.; Diamond, M. L.; Archbold, J. A.; Ferguson, G.; Harner, T.; Richardson, G. M.; Ryan, J. J.; Wilford, B. Is House Dust the Missing Exposure Pathway for PBDEs? An Analysis of the Urban Fate and Human Exposure to PBDEs. *Environ. Sci. Technol.* **2005**, *39*, 5121–5130.

(10) US EPA, O. of P. P. and T. Q. & A. Consumer Fact Sheet on Flame Retardants | Design for the Environment | US EPA http://www.epa.gov/dfe/pubs/projects/flameret/flame-retardants-factsheet.htm (accessed Aug 9, 2014).

(11) Quirós-Alcalá, L.; Bradman, A.; Nishioka, M.; Harnly, M. E.; Hubbard, A.; McKone, T. E.; Eskenazi, B. Concentrations and loadings of polybrominated diphenyl ethers in dust from low-income households in California. *Environ. Int.* **2011**, *37*, 592–596.

(12) Whitehead, T. P.; Brown, F. R.; Metayer, C.; Park, J.-S.; Does, M.; Petreas, M. X.; Buffler, P. A.; Rappaport, S. M. Polybrominated diphenyl ethers in residential dust: Sources of variability. *Environ. Int.* **2013**, *57–58*, 11–24.

(13) Stapleton, H. M.; Dodder, N. G. Photodegradation of decabromodiphenyl ether in house dust by natural sunlight. *Environ. Toxicol. Chem.* **2008**, *27*, 306–312.

(14) US EPA, O. of P. P. and T. Flame Retardants Used in Flexible Polyurethane Foam | Design for the Environment (DfE) | US EPA http://www.epa.gov/dfe/pubs/projects/flameret/about.htm#regulatory (accessed Aug 9, 2014).

(15) O'Connell, S. G.; Kincl, L. D.; Anderson, K. A. Silicone Wristbands as Personal Passive Samplers. *Environ. Sci. Technol.* **2014**, *48*, 3327–3335.
(16) Zota, A. R.; Park, J.-S.; Wang, Y.; Petreas, M.; Zoeller, R. T.; Woodruff, T. J. Polybrominated Diphenyl Ethers, Hydroxylated Polybrominated Diphenyl Ethers, and Measures of Thyroid Function in Second Trimester Pregnant Women in California. *Environ. Sci. Technol.* **2011**, *45*, 7896–7905.
(17) Schecter, A.; Päpke, O.; Joseph, J. E.; Tung, K.-C. Polybrominated Diphenyl Ethers (PBDEs) In U.S. Computers and Domestic Carpet Vacuuming: Possible Sources of Human Exposure. *J. Toxicol. Environ. Health A* **2005**, *68*, 501–513.
(18) Flame Retardants in Furniture. *Green Science Policy Institute*.
(19) Eskenazi, B.; Chevrier, J.; Rauch, S. A.; Kogut, K.; Harley, K. G.; Johnson, C.; Trujillo, C.; Sjodin, A.; Bradman, A. In Utero and Childhood Polybrominated Diphenyl Ether (PBDE) Exposures and Neurodevelopment in the CHAMACOS Study. *Environ. Health Perspect.* **2013**, *121*, 257–262.

3.1 Population Information

Characteristic		Raw			Collapsed	
		n	%		n	%
Mattress						
	Foam	15	19%	Foam	15	19%
	Spring	49	64%	Spring	49	64%
	Mix	3	4%	Mix+Unsure	13	17%
	Unsure	10	13%			
Pillow						
	Foam	2	3%	Foam+Synthetic	41	54%
	Synthetic	39	51%	Down	3	4%
	Down	3	4%	Mix	14	18%
	Mix	14	18%	Unsure	18	24%
	Unsure	18	24%			
Floor Type						
	Carpet	5	7%			
	Wood	5	7%			
	Mix	66	87%			
Carpet Percentage						
	0-25%	19	25%			
	26-50%	16	21%			
	51-75%	11	14%			
	76-100%	31	40%			
Square Footage						
	0-1000sf	13	19%			
	1101-1500sf	19	28%			

	1501-2000sf	15	22%			
	2001-2500sf	12	17%			
	2500+sf	10	14%			
House Built						
	Pre 1975	16	29%			
	1976-2005	26	46%			
	2006-present	11	20%			
Vacuum						
	1-4 times/month	37	48%			
	5-8 times/month	25	32%			
	9+ times/month	15	19%			
Mop						
	0-4 times/month	53	71%	0-4 times/month	53	71%
	5-8 times/month	17	23%	5+ times/month	22	29%
	9+ times/month	5	7%			
Income						
	Less than $22,000	17	24%	Less than $22,000	17	24%
	$22,001-$30,000	3	4%	$22,001-$70,000	25	36%
	$30,001-$38,000	0	0%	$70,001 or more	28	40%
	$38,001-$46,000	4	6%			
	$46,001-$54,000	6	9%			
	$54,001-$62,000	4	6%			
	$62,001-$70,000	8	11%			
	$70,001 or more	28	40%			

(Figure 1)

3.2 Mattress

ANOVA						
		Sum of Squares	df	Mean Square	F	Sig.
TCEP	Between Groups	.764	2	.382	.584	.560
	Within Groups	48.430	74	.654		
	Total	49.194	76			
TCPP	Between Groups	1.907	2	.954	.655	.522
	Within Groups	107.724	74	1.456		
	Total	109.631	76			
TDCPP	Between Groups	1.179	2	.589	.606	.548
	Within Groups	72.019	74	.973		
	Total	73.198	76			
TPP	Between Groups	.608	2	.304	.197	.822
	Within Groups	114.288	74	1.544		
	Total	114.897	76			
PBDE47	Between Groups	1.000	2	.500	.227	.797
	Within Groups	162.745	74	2.199		
	Total	163.745	76			
PBDE100	Between Groups	.942	2	.471	.246	.782
	Within Groups	141.479	74	1.912		
	Total	142.421	76			
PBDE99	Between Groups	.562	2	.281	.113	.893
	Within Groups	184.122	74	2.488		
	Total	184.685	76			
PBDE154	Between Groups	1.657	2	.828	.552	.578
	Within Groups	110.953	74	1.499		
	Total	112.610	76			
PBDE153	Between Groups	3.844	2	1.922	1.138	.326
	Within Groups	124.945	74	1.688		
	Total	128.789	76			

(Figure 2)

3.3 Pillows

ANOVA						
		Sum of Squares	df	Mean Square	F	Sig.
TCEP	Between Groups	4.291	3	1.430	2.325	.082
	Within Groups	44.903	73	.615		
	Total	49.194	76			
TCPP	Between Groups	9.094	3	3.031	2.201	.095
	Within Groups	100.536	73	1.377		
	Total	109.631	76			
TDCPP	Between Groups	1.263	3	.421	.427	.734
	Within Groups	71.935	73	.985		
	Total	73.198	76			
TPP	Between Groups	5.937	3	1.979	1.326	.273
	Within Groups	108.960	73	1.493		
	Total	114.897	76			
PBDE47	Between Groups	10.272	3	3.424	1.629	.190
	Within Groups	153.472	73	2.102		
	Total	163.745	76			
PBDE100	Between Groups	8.946	3	2.982	1.631	.190
	Within Groups	133.475	73	1.828		
	Total	142.421	76			
PBDE99	Between Groups	10.194	3	3.398	1.422	.243
	Within Groups	174.490	73	2.390		
	Total	184.685	76			
PBDE154	Between Groups	5.562	3	1.854	1.264	.293
	Within Groups	107.048	73	1.466		
	Total	112.610	76			
PBDE153	Between Groups	6.996	3	2.332	1.398	.250
	Within Groups	121.793	73	1.668		
	Total	128.789	76			

(Figure 3)

3.4 Comparison of Floor Types

		Sum of Squares	df	Mean Square	F	Sig.
TCEP	Between Groups	.809	2	.405	.611	.546
	Within Groups	48.378	73	.663		
	Total	49.187	75			
TCPP	Between Groups	4.265	2	2.132	1.482	.234
	Within Groups	105.019	73	1.439		
	Total	109.284	75			
TDCPP	Between Groups	.824	2	.412	.416	.661
	Within Groups	72.306	73	.990		
	Total	73.130	75			
TPP	Between Groups	2.422	2	1.211	.803	.452
	Within Groups	110.160	73	1.509		
	Total	112.582	75			
PBDE47	Between Groups	8.136	2	4.068	1.926	.153
	Within Groups	154.157	73	2.112		
	Total	162.294	75			
PBDE100	Between Groups	8.090	2	4.045	2.228	.115
	Within Groups	132.549	73	1.816		
	Total	140.639	75			
PBDE99	Between Groups	8.815	2	4.407	1.854	.164
	Within Groups	173.571	73	2.378		
	Total	182.386	75			
PBDE154	Between Groups	3.241	2	1.621	1.095	.340
	Within Groups	108.027	73	1.480		
	Total	111.268	75			
PBDE153	Between Groups	3.996	2	1.998	1.184	.312
	Within Groups	123.169	73	1.687		
	Total	127.165	75			

(Figure 4)

3.5 Comparison of Percentage of Home Covered by Carpet

		Sum of Squares	df	Mean Square	F	Sig.
TCEP	Between Groups	.812	3	.271	.408	.748
	Within Groups	48.382	73	.663		
	Total	49.194	76			
TCPP	Between Groups	6.086	3	2.029	1.430	.241
	Within Groups	103.545	73	1.418		
	Total	109.631	76			
TDCPP	Between Groups	1.143	3	.381	.386	.764
	Within Groups	72.055	73	.987		
	Total	73.198	76			
TPP	Between Groups	.168	3	.056	.036	.991
	Within Groups	114.729	73	1.572		
	Total	114.897	76			
PBDE47	Between Groups	.378	3	.126	.056	.982
	Within Groups	163.367	73	2.238		
	Total	163.745	76			
PBDE100	Between Groups	.686	3	.229	.118	.949
	Within Groups	141.735	73	1.942		
	Total	142.421	76			
PBDE99	Between Groups	.361	3	.120	.048	.986
	Within Groups	184.324	73	2.525		
	Total	184.685	76			
PBDE154	Between Groups	1.098	3	.366	.240	.868
	Within Groups	111.512	73	1.528		
	Total	112.610	76			
PBDE153	Between Groups	2.493	3	.831	.480	.697
	Within Groups	126.296	73	1.730		
	Total	128.789	76			

(Figure 5)

3.6 Comparison of Size of House

		Sum of Squares	df	Mean Square	F	Sig.
TCEP	Between Groups	2.873	4	.718	1.079	.375
	Within Groups	42.620	64	.666		
	Total	45.493	68			
TCPP	Between Groups	2.389	4	.597	.373	.827
	Within Groups	102.588	64	1.603		
	Total	104.977	68			
TDCPP	Between Groups	3.903	4	.976	1.022	.403
	Within Groups	61.102	64	.955		
	Total	65.005	68			
TPP	Between Groups	11.217	4	2.804	2.057	.097
	Within Groups	87.249	64	1.363		
	Total	98.466	68			
PBDE47	Between Groups	12.585	4	3.146	1.501	.212
	Within Groups	134.166	64	2.096		
	Total	146.751	68			
PBDE100	Between Groups	7.492	4	1.873	1.035	.396
	Within Groups	115.802	64	1.809		
	Total	123.293	68			
PBDE99	Between Groups	15.141	4	3.785	1.589	.188
	Within Groups	152.423	64	2.382		
	Total	167.564	68			
PBDE154	Between Groups	7.788	4	1.947	1.452	.227
	Within Groups	85.800	64	1.341		
	Total	93.588	68			
PBDE153	Between Groups	4.176	4	1.044	.696	.598
	Within Groups	96.057	64	1.501		
	Total	100.233	68			

(Figure 6)

3.7 Comparison of Age of Home

		Sum of Squares	df	Mean Square	F	Sig.
TCEP	Between Groups	2.128	2	1.064	1.541	.224
	Within Groups	34.520	50	.690		
	Total	36.648	52			
TCPP	Between Groups	3.616	2	1.808	1.236	.299
	Within Groups	73.125	50	1.462		
	Total	76.741	52			
TDCPP	Between Groups	4.241	2	2.120	2.177	.124
	Within Groups	48.707	50	.974		
	Total	52.948	52			
TPP	Between Groups	.400	2	.200	.155	.857
	Within Groups	64.328	50	1.287		
	Total	64.727	52			
PBDE47	Between Groups	2.998	2	1.499	.783	.462
	Within Groups	95.669	50	1.913		
	Total	98.667	52			
PBDE100	Between Groups	3.109	2	1.555	.908	.410
	Within Groups	85.556	50	1.711		
	Total	88.665	52			
PBDE99	Between Groups	6.627	2	3.314	1.607	.211
	Within Groups	103.125	50	2.063		
	Total	109.753	52			
PBDE154	Between Groups	1.582	2	.791	.619	.542
	Within Groups	63.864	50	1.277		
	Total	65.446	52			
PBDE153	Between Groups	1.003	2	.501	.333	.718
	Within Groups	75.308	50	1.506		
	Total	76.311	52			

(Figure 7)

.

3.8 Comparison of Monthly Vacuuming

		Sum of Squares	df	Mean Square	F	Sig.
TCEP	Between Groups	1.384	2	.692	1.071	.348
	Within Groups	47.810	74	.646		
	Total	49.194	76			
TCPP	Between Groups	6.343	2	3.171	2.272	.110
	Within Groups	103.288	74	1.396		
	Total	109.631	76			
TDCPP	Between Groups	.500	2	.250	.255	.776
	Within Groups	72.697	74	.982		
	Total	73.198	76			
TPP	Between Groups	2.373	2	1.187	.780	.462
	Within Groups	112.524	74	1.521		
	Total	114.897	76			
PBDE47	Between Groups	9.566	2	4.783	2.296	.108
	Within Groups	154.179	74	2.084		
	Total	163.745	76			
PBDE100	Between Groups	4.776	2	2.388	1.284	.283
	Within Groups	137.645	74	1.860		
	Total	142.421	76			
PBDE99	Between Groups	13.106	2	6.553	2.826	.066
	Within Groups	171.579	74	2.319		
	Total	184.685	76			
PBDE154	Between Groups	10.394	2	5.197	3.762	.028
	Within Groups	102.217	74	1.381		
	Total	112.610	76			
PBDE153	Between Groups	14.139	2	7.070	4.563	.014
	Within Groups	114.650	74	1.549		
	Total	128.789	76			

(Figure 8)

3.9 Comparison of Monthly Mopping

Independent Samples Test

		Equality of Variances		t-test for Equality of Means						
									95% Confidence	
		F	Sig.	t	df	Sig. (2-tailed)	Mean Difference	Std. Error Difference	Lower	Upper
TCEP	Equal variances assumed	.307	.581	-.676	73	.501	-.1402514	.2075284	-.5538550	.2733521
	Equal variances not assumed			-.667	38.256	.509	-.1402514	.2101801	-.5656454	.2851425
TCPP	Equal variances assumed	.681	.412	1.070	73	.288	.3285821	.3070445	-.2833566	.9405209
	Equal variances not assumed			1.111	42.800	.273	.3285821	.2956839	-.2678018	.9249660
TDCPP	Equal variances assumed	.716	.400	-.816	73	.417	-.2057684	.2522265	-.7084551	.2969184
	Equal variances not assumed			-.915	51.611	.364	-.2057684	.2248900	-.6571240	.2455873
TPP	Equal variances assumed	1.359	.248	-.750	73	.456	-.2343131	.3125743	-.8572729	.3886466
	Equal variances not assumed			-.672	31.710	.507	-.2343131	.3488543	-.9451608	.4765345
PBDE47	Equal variances assumed	.751	.389	-.792	73	.431	-.2981336	.3763366	########	.4519042
	Equal variances not assumed			-.876	49.813	.385	-.2981336	.3403484	-.9818069	.3855397
PBDE100	Equal variances assumed	.553	.459	-.667	73	.507	-.2339991	.3509537	-.9334488	.4654506
	Equal variances not assumed			-.682	41.364	.499	-.2339991	.3430321	-.9265813	.4585831
PBDE99	Equal variances assumed	.002	.969	-.305	73	.761	-.1222909	.4006145	-.9207144	.6761327
	Equal variances not assumed			-.311	40.905	.758	-.1222909	.3935108	-.9170578	.6724761
PBDE154	Equal variances assumed	.030	.863	-.787	73	.434	-.2452744	.3115809	-.8662542	.3757053
	Equal variances not assumed			-.776	38.088	.443	-.2452744	.3162159	-.8853714	.3948225
PBDE153	Equal variances assumed	.038	.846	-.813	73	.419	-.2708083	.3331805	-.9348361	.3932195
	Equal variances not assumed			-.801	38.063	.428	-.2708083	.3382431	-.9555087	.4138921

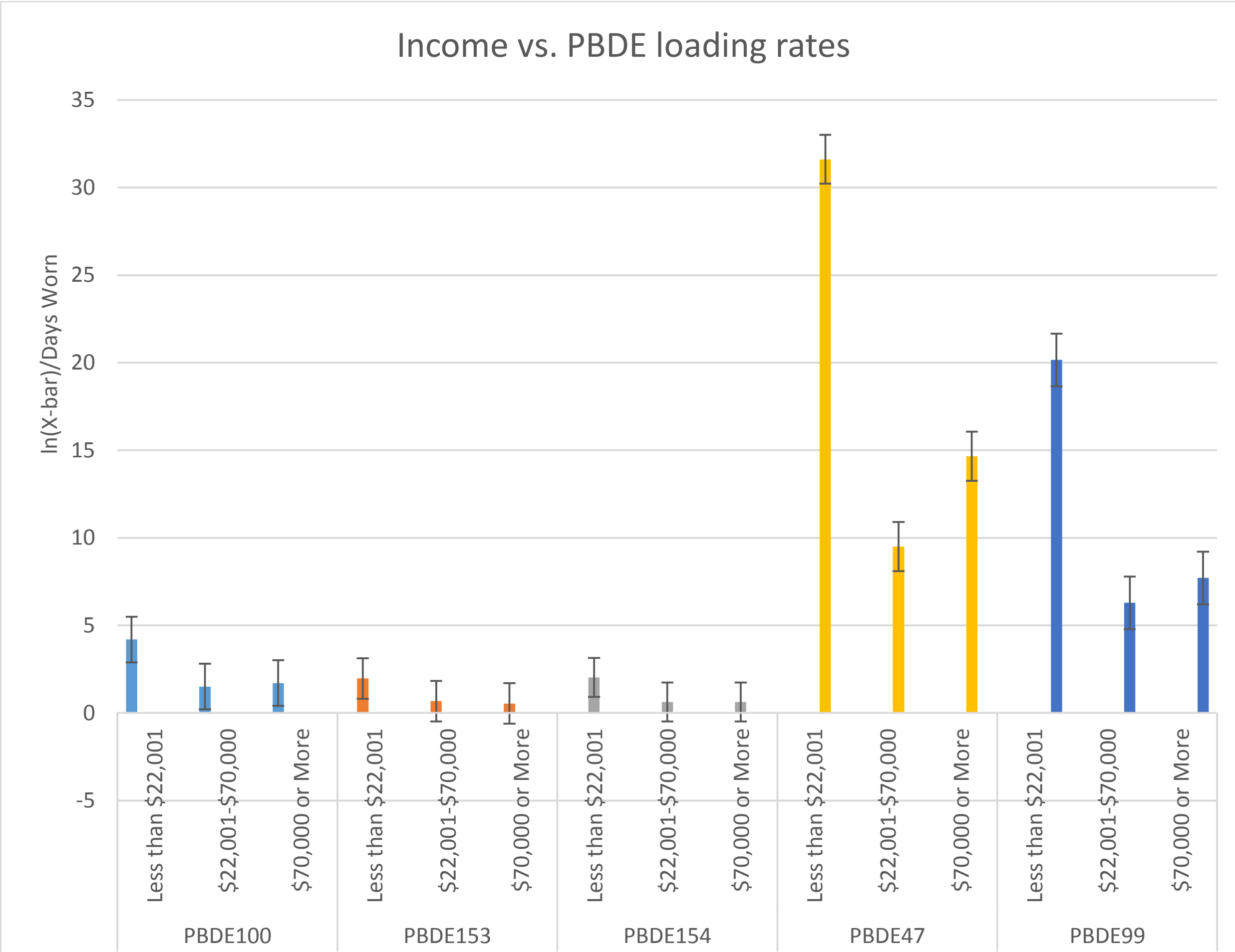
(Figure 9)

3.10 Comparison of Household Income

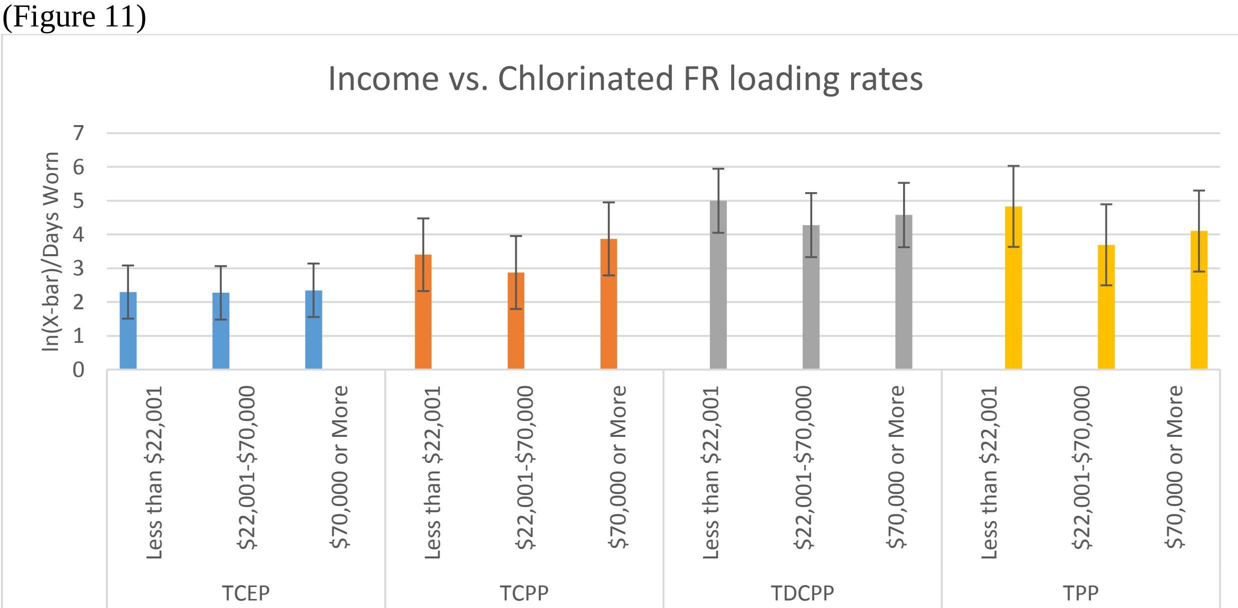
		Sum of Squares	df	Mean Square	F	Sig.
TCEP	Between Groups	.084	2	.042	.063	.939
	Within Groups	49.110	74	.664		
	Total	49.194	76			
TCPP	Between Groups	14.766	2	7.383	5.759	.005
	Within Groups	94.865	74	1.282		
	Total	109.631	76			
TDCPP	Between Groups	5.769	2	2.885	3.166	.048
	Within Groups	67.428	74	.911		
	Total	73.198	76			
TPP	Between Groups	14.438	2	7.219	5.318	.007
	Within Groups	100.459	74	1.358		
	Total	114.897	76			
PBDE47	Between Groups	16.061	2	8.030	4.024	.022
	Within Groups	147.684	74	1.996		
	Total	163.745	76			
PBDE100	Between Groups	12.556	2	6.278	3.577	.033
	Within Groups	129.865	74	1.755		
	Total	142.421	76			
PBDE99	Between Groups	15.780	2	7.890	3.457	.037
	Within Groups	168.904	74	2.282		
	Total	184.685	76			
PBDE154	Between Groups	17.960	2	8.980	7.021	.002
	Within Groups	94.650	74	1.279		
	Total	112.610	76			
PBDE153	Between Groups	18.941	2	9.470	6.380	.003
	Within Groups	109.848	74	1.484		
	Total	128.789	76			

(Figure 10)

3.11 Trends in Income Levels



(Figure 11)



(Figure 12)