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Do flame retardant concentrations change in dust after older upholstered furniture is replaced?

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Abstract

Upholstered furniture has been a major source of chemical flame retardant (FR) exposures in US homes since the 1970s. FRs are a large group of chemicals, many of which are associated with adverse health effects, including cancer, reproductive toxicity, and neurotoxicity. California homes have some of the highest dust concentrations of FRs, due to Technical Bulletin 117 (TB117), California's outdated flammability standard for furniture foam that was generally followed across the US and Canada. In 2014, this standard was updated to a smolder standard for furniture fabric called TB117-2013, and it is no longer reliant on FRs. This update provided an opportunity to measure differences in FR dust levels in California homes before and after residents replaced older upholstered furniture, or its foam, with products that met the new standard and were expected to be FR-free. We collected dust from homes of participants who had plans to replace older upholstered furniture, or furniture foam, with FR-free options. We returned for follow-up dust collection six, 12, and 18 months following replacement. Concentrations of three polybrominated diphenyl ethers (PBDEs) (BDE-47, BDE-99, BDE-100), three chlorinated organophosphate ester

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FRs (tris(2-chloroethyl) phosphate (TCEP), tris(2-chloroisopropyl) phosphate (TCIPP), and tris(1,3-dichloroisopropyl) phosphate (TDCIPP)), and one aryl organophosphate ester FR triphenyl phosphate (TPHP), were widely detected in participant homes. All measured FRs decreased in nearly all homes after the older upholstered furniture was replaced. The decreases in FRs were significant in both homes that replaced entire pieces of furniture and those that replaced only the furniture foam. This study demonstrates that replacing older upholstered furniture or foam significantly reduces concentrations of a range of FRs in the home. Foam replacement offers a potentially more economic alternative that produces a lower volume of waste.

Keywords

flame retardants; exposure reduction; furniture; flammability standard; house dust

1. INTRODUCTION

Prior to 2013, flame retardant (FR) chemicals were added to foam in upholstered furniture to meet a California furniture flammability standard that was written in 1975, called Technical Bulletin (TB) 117. The standard was typically met by adding large quantities of FRs to the foam; one study found FRs at up to five percent by weight in furniture foam that met TB117 (Stapleton et al. 2009). Additive FRs, which are not chemically bound to their substrate, migrate from products like furniture and can settle into house dust (Dodson et al. 2012). Dust is a major exposure pathway for many FRs, with exposures among children being a particular concern due to their proximity to the floor and increased hand-to-mouth activity (Mitro et al. 2016; Wu et al. 2015). In the absence of a national upholstered furniture flammability standard, nationally distributed brands often met TB117, which meant that most upholstered furniture sold in the US until 2014 contained FRs. To improve fire safety, California revised TB117 in 2013 to a smolder standard that furniture manufactures can meet without adding FRs to the foam (State of California 2013, 2014a). This change in legislation allows for the evaluation of FR dust levels following the removal of FR-containing furniture or furniture foam from homes.

FRs are a topic of concern for public health because they are a large group of chemicals associated with many adverse health effects, including cancer, neurotoxicity, reproductive and developmental toxicity (Blum et al. 2019; Kim et al. 2014; Wu et al. 2020). PentaBDE was the commercial mixture of polybrominated diphenyl ethers (PBDEs) used in upholstered furniture for decades, until they were phased out in 2004 (Dodson et al. 2012; Stapleton et al. 2012). They are associated with decreased IQ (Lam et al. 2017), due to their ability to disrupt thyroid hormones necessary for gene signaling and myelination during brain development (Bernal 2007; Dorman et al. 2018). PBDEs have also been found to disrupt hormone signaling in cell studies (Mercado-Feliciano and Bigsby 2008) and are associated with thyroid disease in women (Allen et al. 2016). A health cost analysis estimated PBDEs led to $166 billion in disease costs in the US in 2010, largely due to lost IQ (Attina et al. 2016). Tris(1,3-dichloroisopropyl) phosphate (TDCIPP or chlorinated “tris”) was removed from children’s pajamas in 1978 after toxicity testing found its metabolites were mutagenic (Gold et al. 1978); however, it continued to be used in furniture and other consumer applications. TDCIPP is listed as a carcinogen on California's Proposition 65 (California Office of Environmental Health Hazard Assessment 2020) and has also shown evidence of neurotoxicity in experimental studies (Dishaw et al. 2011). Other chlorinated tris compounds such as tris(2-chloroethyl) phosphate (TCEP) and tris(2-chloroisopropyl) phosphate (TCIPP) have also been used in furniture and are of concern. TCEP is also listed as carcinogen on California's Proposition 65 (California Office of Environmental Health Hazard Assessment 2020) and is a Substance of Very High Concern and is considered persistent, bioaccumulative and toxic (PBT) under REACH (ECHA 2009). Emerging evidence on organophosphate ester FRs (OPFRs) shows associations with impaired learning, reproductive problems, asthma and allergy, and behavioral problems in children (Araki et al. 2018; Blum et al. 2019; Carignan et al. 2018; Castorina et al. 2017; Ingle et al. 2018; Lipscomb et al. 2017; Meeker and Stapleton 2010). Regulations restricting the use of TCEP and TDCIPP in furniture and some consumer products have been issued by some states because of health concerns (California Department of Toxic Substances Control 2017; Safer States 2021).

Studies have shown that presence of PBDEs in furniture foam and dust in the home were associated with resident serum levels of PBDEs (Hammel et al. 2017; Johnson et al. 2010), and it has been shown that foam recyclers and carpet installers who handled foam had PBDE serum levels that were higher than the general population (Stapleton et al. 2008). Prior studies have found that behavioral interventions, such as more frequent hand washing, can reduce human body burdens of PBDEs (Watkins et al. 2011). A study that prescribed one week of enhanced cleaning in homes, including vacuuming with a HEPA filter, found lower urinary levels of a TCIPP metabolite and a triphenyl phosphate (TPHP) metabolite in home occupants (Gibson et al. 2019). Studies have shown that when FRs are removed from indoor furnishings, either through manufacturing phase outs or furniture standard changes, the dust and blood levels of those chemicals decline (Dodson et al. 2012; Parry et al. 2018; Rodgers et al. 2020; Stubbings et al. 2018).

After 2013, for the first time in nearly 40 years, people in California had the option to purchase furniture or furniture foam free of FR chemicals. A separate 2015 California labeling law also required a label on new furniture to indicate if FRs were added to furniture or not (State of California 2014b), and an additional law passed in 2018 banned FRs from upholstered furniture sold in California at levels above 1000 parts per million (ppm) due to the health effects associated with flame retardant exposures (State of California 2018). The goal of our study was to quantify the effect of replacing older upholstered furniture or furniture foam, which likely contained FRs, with furniture or furniture foam manufactured to meet TB117-2013, and therefore expected to be FR-free, on levels of FRs in house dust. We recruited participants from Northern California who were replacing either the older upholstered furniture or furniture's seating foam in their main living area. In this paper we analyzed the changes in FR dust concentrations for seven FRs in homes that had measurements taken before the furniture replacement and six, 12, and 18 months after replacement.

2. MATERIALS AND METHODS

2.1 Study population

Participants were recruited through two different efforts. The first effort recruited individuals who were voluntarily planning on replacing either their older upholstered furniture or the foam in their furniture, and were willing to participate in a study that measured dust concentrations before and after replacement, which we refer to as the “voluntary replacement” group. The second effort recruited individuals to take part in a furniture replacement program, where the replacement furniture was purchased for them, which we refer to as the “purchased replacement“ group. All enrolled homes were in Northern California.

Twenty-eight participants in the voluntary replacement group were recruited jointly by researchers at the Green Science Policy Institute (GSPI) and the University of California, Davis (UC Davis). Invitations to join the study were sent to the GSPI mailing list, which included people participating in their program facilitating foam replacement, called the Safer Sofa Foam Exchange program (Green Science Policy Institute 2019). Those on the mailing list, as well as GSPI and UC Davis study staff, were also encouraged to distribute the study invitation to others they thought would be interested. To be eligible for the study, participants’ homes were required to have at least one couch or loveseat in the main living area that was manufactured after 1975 and before 2014, thus presumed to contain added FRs. Two exceptions were made for participants who had antique couches with foam that had been replaced in the 2000s. Participants needed to plan to replace the foam in the identified item, or replace the item altogether, with furniture or foam thought to be FR-free within one year from the start of the study. If a participant had two equal sized couches or loveseats in the main living area, they were required to replace both, unless the second was already FR-free. Large overstuffed or foam filled chairs were also required to be replaced. Smaller upholstered furniture items, such as dining room chairs or ottomans, were allowed to remain. For participants in the voluntary replacement group, new furniture was checked for TB117-2013 tags or other tags indicating whether FRs were added or not at the 6-month visit. In addition, only participants not planning to move in the next two years were eligible for the study.

All fourteen participants in the purchased replacement group lived in one of two affordable housing complexes. GSPI organized a furniture replacement program that provided new furniture for the participating households. Flyers were distributed to the residents, and a community meeting was held at each complex to inform residents of the study and recruit those interested. Eligibility requirements were the same as the voluntary replacement group. However, in this group, participants selected replacement furniture from a set of options from a retailer that provided assurances that the furniture did not contain added FRs. All furniture was checked for TB117-2013 tags before the furniture was installed in the homes. All recruitment and data collection protocols were approved by the Institutional Review Board for UC Davis. Participants provided informed consent before collection of any data.

2.2 Sample Collection

Trained study staff vacuumed the main living space in participants' homes to collect dust for FR analysis before furniture or foam replacement and again six, 12, and 18 months after the furniture or foam was replaced. A standardized protocol was used to vacuum all surfaces except for under furniture and between cushions (Allen et al. 2008; Rudel et al. 2003), with modifications to also exclude vacuuming upholstered furniture. Dust samples were collected with a Eureka Mighty-Mite vacuum cleaner equipped with the standard crevice tool attachment (Model 3670), modified to capture dust in a 19×90 mm cellulose extraction thimble (Whatman Inc.) (Allen et al. 2008; Rudel et al. 2003). Cleaned crevice tools were used in each home. Crevice tools were cleaned with a 1% Alconox detergent solution in advance of sample collection visits, following a standard protocol. The thimbles containing the dust samples were wrapped in pre-cleaned aluminum foil, placed in zip-top bags, transported to UC Davis in a cooler with blue ice, and stored at −20 °C until analysis. In the volunteer replacement group, a prereplacement dust sample was collected from July 2015 to September 2016. In the purchased replacement group, pre-replacement dust was collected from all homes in May 2016.

In order to identify which FRs were removed from the home, participants in the voluntary replacement group were asked if we could take an approximately 1-inch cube foam sample from the furniture being replaced, noting that this was generally only practical if the foam was accessible by a zipper since participants continued to use the couch until they replaced it, and thus only some of the participants agreed. For the purchased replacement group, study staff were on hand the day the furniture was replaced and were thus able to collect samples from the seat foam, arm foam, decking material, and fabric from the items removed. To determine if the new furniture contained FRs, since foam manufacturing practices were not uniform in the initial months following the furniture standard change, we collected a piece of foam from new furniture items in the voluntary replacement group if the participant was willing. As all furniture in the purchased replacement group came from the same furniture manufacturer and was checked for TB117-2013 tags at installation, four foam samples from this group were collected to effectively represent all the furniture in this group.

2.3 Questionnaire

Study staff collected detailed information on the home during sample collection visits through an inperson questionnaire, including the square footage of living room, floor type, carpeting and rugs, frequency of vacuuming, if the home used a vacuum with a HEPA filter, and the number of people living in the home. For each upholstered furniture item in the main living area observed at the initial study visit, study staff recorded if participants planned to replace or remove the entire furniture item, replace the foam in the item, or in the case of a "small" item, if it would be kept in the same room or moved to another area of the home. At each follow-up visit the status (replace/removed, foam replaced, new location in home, or still in room) of each furniture item from the main living area was recorded. We applied a weighting scheme for the furniture items (couch=1, loveseat=0.75, chair=0.5, ottoman=0.25, double sectional=2) to represent the approximate relative volume of foam in the various items. At each visit, we also collected detailed information about the furniture, electronics, and baby items with foam (e.g. high chairs) in the main living area as well as adjoining rooms. We also collected demographic characteristics, including race/ethnicity, education status, and income, of the study participants.

2.4 Dust analysis

Extraction and analysis of dust followed methods previously reported (Moschet et al. 2018). Dust samples were sieved (106 μm) and sieved samples (100 mg) were extracted with hexane/acetone (3:1 v/v) followed by acetone (100%) by sonication. The combined supernatant was evaporated under nitrogen using a Turbovap (Biotage) and filtered through a PTFE filter (0.2 μm). Dust samples and extracts were stored frozen (−20 °C) except when being analyzed. Samples were analyzed in three batches, with all the dust from pre-replacement and six-month collection times extracted and analyzed in one batch, and the majority of the dust samples from the 12 and 18 month samples analyzed in a second batch, with a small number analyzed in a third batch; storage time before extraction ranged from 0 days to 21 months.

We quantified concentrations of 13 FRs, specifically seven BDEs (BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, and BDE-183), three chlorinated OPFRs (TCEP, TCIPP, and TDCIPP), and three alkyl and aryl OPFRs (tris(2-butoxyethyl) phosphate (TBOEP), triphenyl phosphate (TPHP), and tri-n-butyl-phosphate (TNBP)). Analysis was carried out on an Agilent 7890B gas chromatograph with an HP-5MS (30 m × 0.25 mm, 0.25 μm) column coupled to an Agilent Q/TOF 7200B instrument running in electron ionization (EI) mode. A 78 min. run time with a linear temperature gradient from 35 to 325 °C was chosen to separate all major peaks in the analysis of a standard reference house dust (NIST SRM 2585).

2.5 Furniture foam and textile analysis

Extraction and analysis of foam and textile samples followed methods previously reported (California Environmental Protection Agency 2019a, b; Petreas et al. 2016). Foam and/or textile samples were homogenized by cryogenic milling when sufficient sample material was available. Homogenized samples (50 mg) were extracted in toluene (100%) by sonication. The extraction process was repeated twice and the supernatant filtered through a set of PE filters (20/10 μm). Foam and/or fabric samples were stored at <6°C except when being analyzed. New foam (thought to be FR-free) was prepared and carried through the complete sample preparation and extraction process. Each sample preparation and analysis batch included the following quality control samples: method blank, laboratory control sample, laboratory control duplicate, reference foam material (7.5 wt% TCEP), and a sample duplicate.

We quantified concentrations of 17 FRs in foam and/or textile samples: nine PBDEs (BDE-17, BDE-28, BDE-47, BDE-85, BDE-99, BDE-100, BDE-153, BDE-154 and BDE-183), six OPFRs (tri-propyl phosphate (TPP), TNBP, TCEP, TCIPP, TDCIPP and TPHP) and two BFRs (2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB) and bis(2-ethylhexyl)-2,3,4,5-tetrabromophthalate (BEH-TEBP)). Table S1 list the CAS numbers and corresponding internal standards. Analysis was performed on an Agilent 7890B gas chromatograph with a DB-5MS (30 m × 0.25 mm, 0.25 μm film) column coupled to an

Agilent 7000 series electron ionization (EI) triple quadrupole mass spectrometer (GC-MS/MS) operated in multiple reaction monitoring (MRM) mode.

2.6 Quality Assurance/Quality Control

We used multiple QA/QC measures to ensure we reported accurate and precise FR concentrations in dust. Four method blanks were analyzed in the first batch (pre-replacement and six month) and two were included in the second batch (12 and 18 month). If analytes were detected in more than half of the blank samples, then we blank corrected those analytes' raw concentrations. BDE-47, TCIPP, TDCIPP, and TPHP were blank corrected by subtracting the median blank values (in ng/g) from the raw concentrations. We used machine-read values (values below the limit of quantification (LOQ) but above the limit of detection (LOD)) in subsequent statistical analyses, and we set samples that were below the LOQ to the LOD in subsequent statistical analyses.

The LOQ for each analyte was calculated as 10 times the baseline values from the chromatogram; the LOD was calculated as three times the baseline values. The LOD was blank corrected by subtracting the peak areas of the corresponding analytes in the method blanks. If the blank corrected LOD was higher than the baseline-derived LOQ, then the LOQ was adjusted to match the blank corrected LOD.

The accuracy of six PBDEs was estimated for batches 1 and 2, and was calculated using certified values. Accuracy ranged between 68-90% for the PBDEs included in subsequent data analyses. See supplementary material for additional details on quality assurance/quality control methods.

2.7 Data analysis

Of the thirteen FRs analyzed in dust, we prioritized chemicals for data analysis if they were detected in any foam or fabric samples, and if they were above the maximum LOQ in at least 50% of samples in at least one time period (detection limits varied between batches). Seven chemicals were prioritized for data analysis: three PBDEs used in the PentaBDE mixture that was known to be applied to polyurethane foam in furniture (BDE-47, BDE-99, and BDE-100), three chlorinated OPFRs (TCEP, TCIPP, and TDCIPP), and one aryl OPFR (TPHP). We summarized FR concentrations and questionnaire data by study visit, type of furniture replacement, and replacement group (voluntary versus purchased) using descriptive statistics and visualizations.

To evaluate the effect of the furniture replacement on FRs in dust concentrations, we used linear mixed effects models for our primary analysis, with each household as a random effect, which allows for individual effects of each house, and the two time points, six and 12 months, after the replacement as fixed effects. Within each model, we evaluated the short-term effect using a binary time variable (t1=1 for six-month data and t1=0 if not six-month data) and longer-term effect using a second binary time variable (t2=1 for 12-month data and t2=0 if not 12-month data). As a sensitivity analysis, we reestimated our models using the 18-month data in place of the 12-month data. We log-transformed concentrations (the outcome) for modeling. We also ran these models stratified by foam versus furniture replacement in order to look for differences in effect by the type of replacement.

We were also interested in a priori identified variables relevant to the effect of furniture replacement on FR concentrations. Because of limited statistical power to include these variables in our mixed effects models described above, we estimated the mixed effects models stratified by these variables of interest. We stratified analyses by use of HEPA vacuum, vacuum frequency, carpeting status, square footage of the living area, number of people living in the home, weighted percent of furniture replaced, and weighted furniture density of the main living area. In the case of continuous variables, such as square foot of the living room, were split the data into high and low categories based on the median value. We visualized the changes in FR concentrations for each stratum of these a priori variables, noting significant changes in concentrations from pre-replacement concentrations. We did not expect the replacement of FR-containing furniture and associated changes in FR levels in dust to vary by socioeconomic status (SES), even if SES may be reflected in different items in the home. Setting each household as a random effect in our model effectively controls for this by allowing for individual variation within each home.

In a secondary analysis, we limited our dataset to homes that had their old furniture foam analyzed for FRs. For each FR, we visualized concentrations before and after furniture or foam replacement in homes that removed a source of that FR from the home. The smaller dataset was not sufficiently large for statistical comparisons; however, visualizations of this smaller dataset provided a way to discern the effect of removing specific FRs from a space on dust levels of that FR.

Analyses were conducted in R version 3.6.0 (version 3.6.0; R Development Core Team).

2.8 Study results reporting

We provided personal results reports to participants who consented to receive them. Participants used unique passcodes to access their report online via the Digital Exposure Report-Back Interface (DERBI) (Boronow et al. 2017). We mailed paper reports to participants without email addresses. Each report summarized a household’s change in FR levels after the furniture intervention, how their results compared to other study participants, and FRs that were detected at or above 1000 parts per million in their pre-replacement furniture samples. Reports included an overview of study findings, what is known about health effects, strategies for reducing exposure, and contact information to speak with a researcher. All participants consented to receive their results.

3. RESULTS

3.1 Participant and household characteristics

In total, 42 participants consented to the study; 28 were from the voluntary replacement group, with 22 meeting eligibility requirements by replacing furniture (n=9) or foam (n=13), and 14 were from the purchased replacement group, with 12 replacing furniture. Participants dropped out over the course of the study either because they moved or became unreachable. Retention was higher in the voluntary replacement group, with 21 of the 22 homes completing all four study visits. Of the 12 homes in the purchased replacement group, 7 completed all four study visits (Figure S1). Of the 22 voluntary replacement participants, 12 permitted collection of a pre-replacement foam sample, while we were able to collect foam and fabric from 11 of the 12 purchased replacement group (Figure 1 and Figure S1).

The majority of our study population that completed at least one follow-up visit had a graduate degree and vacuumed with a HEPA filter, although these observations were driven by the voluntary replacement group (Table 1). A higher percentage of households in the purchased replacement group vacuumed at least once a week. About half of all participants replaced at least 90% of furniture (weighted) in their main living area. All participants in the purchased replacement group had installed wall-to-wall carpet, whereas 14% of participants in the voluntary replacement group had installed carpet, with most having hard floors with area rugs. The median square footage of the main living areas in purchased replacement group was smaller than voluntary replacement group.

In the voluntary replacement group, the time between the baseline sampling and when the furniture was replaced ranged from 18 days to 1 year, with a median of 2.8 months. In the purchased replacement group, all pre-replacement samples were collected in May 2016 and all couches were replaced in July 2016.

3.2 Dust concentrations

We detected all seven prioritized FRs (Table 2). TCIPP and TDCIPP were detected in 100% of samples in all four study visits. Detection frequencies for TPHP were 100% in the first two study visits, but were lower (75%) in the later two visits. Of PBDEs, BDE-47 and BDE-99 were found most frequently across all study visits, while BDE-100 was detected in roughly half of samples taken after the first visit. Concentration distributions for all FRs were positively (right) skewed with 95th percentiles up to 12× the median (BDE-47). Concentrations of TCIPP and TDCIPP were among the highest, with TCIPP found at the highest concentration, with a maximum concentration of 280,000 ng/g measured six months after replacement.

3.3 Flame retardants in foam and fabric samples

We collected foam and, in the case of the purchased replacement group, foam and fabric samples, from old furniture from 23 participants who replaced their foam or furniture. From some of these homes, we collected samples from multiple pieces of furniture. Of the FRs that were also measured in dust, TPHP was the most commonly detected FR greater than or equal to 1000 ppm, found in 17 of 23 homes that had old furniture samples tested, followed by TDCIPP (in 8 homes) and PBDEs (in 5 homes) (Figure 1). Often, there were common combinations of FRs found in the furniture, including TPHP, EH-TBB and BEH-TEBP, found in furniture in 13 homes, and TPHP with a suite of BDE compounds found in furniture in five homes (Figure 1). This is not surprising, given that TPHP, EH-TBB, and BEH-TEBP were used in a commonly used mixture called Firemaster 550 (Phillips et al. 2017). Generally, the fabric had much lower levels of FRs than the foam, and typically never had levels greater than trace concentrations of compounds not found in the foam (Figure S10).

Of the nine participants in the voluntary replacement group who removed furniture, five of the new items specifically had tags that said no FRs were added, and foam was collected from one of these items. Furniture in three homes did not have tags, because they either came with no tag or because participants may have removed them, and foam was collected from all of these items. The remaining home selected an item without foam. Of the 13 homes replacing foam, 10 had foam samples collected. Because the purchased recruitment group all received furniture from the same furniture manufacturer, only four foam samples were collected. Of the new foam samples, three were heterogeneous, rather than homogenous foam, consisting of shredded or re-bonded foam, all of which had non-detectable levels of FR. All foam samples from new furniture had non-detectable or trace levels of FRs (Figure 1).

3.4 Impact of furniture replacement on dust concentrations

In participating homes, FR concentrations clearly declined six months after furniture or foam replacement, with a few exceptions (Figure 2). In 10 of the 13 foam replacement homes and in 15 of the 20 furniture replacement homes, all three PBDE and all three chlorinated OPFR concentrations were lower six months after replacement. TPHP concentrations decreased in eight foam replacement homes and 19 furniture replacement homes six months after replacement. PBDE concentrations in the home with the highest pre-replacement concentrations dropped by an average factor of 12.

In our primary mixed effects model analyses, dust concentrations decreased between pre-replacement and post-replacement study visits for all FRs (Table 3). Changes in FR concentrations were generally similar and in the same direction in homes that replaced their furniture's foam and the homes that replaced their entire piece of furniture (Table 3; Figure S9).

Replacement was associated with reductions of all PBDE levels in the study. The geometric means of BDE-47, BDE-99, and BDE-100 decreased by roughly 50% from the pre-replacement to the six-month measurement across both replacement types; all decreases were significant. Decreases, while still significant, were not as large between pre-replacement and 12-month concentrations. When we stratified by replacement type, reductions in BDE-47 and BDE-99 concentrations were larger in the foam replacement versus the furniture replacement groups. Reductions were similar, if not slightly lower, in the foam replacement group for BDE-100. Our sensitivity analysis found that changes at 18 months were similar to the 12-month results (Table S4).

Geometric means of TCEP, TCIPP, and TDCIPP significantly decreased between pre-replacement to six months and pre-replacement to 12 months, with larger declines between pre-replacement and six months. At six months, geometric means decreased between 43% and 51% for the chlorinated OPFRs, and at 12 months, decreases were smaller and ranged between 21% and 31%. In models stratified by replacement type, we also saw larger declines between pre-replacement and six months versus prereplacement and 12 months. Between pre-replacement and six months, TCEP and TCIPP declines were larger in the foam replacement group versus the furniture replacement group, whereas TDCIPP declines were larger in the furniture replacement group compared to the foam replacement group. Of the chlorinated OPFRs, the only significant decrease at 12 months was for TCEP in the foam replacement group. The results at 18 months were similar to the 12-month results, with the one exception that the decrease in TDCIPP at 18 months was no longer significantly different than zero (Table S4). It is possible that this change is due to reduced statistical power, because there were four fewer homes in the study at the 18-month visit, or due to unaccounted for TDCIPP-containing items in the homes.

TPHP was the only FR that decreased to a greater extent between pre-replacement and 12 months (71% decrease) than between pre-replacement and six months (39% decrease). Both reductions in concentrations were significant. This pattern of greater decreases at 12 months than at six months was found in both the furniture and foam replacement groups. The 12 month and 18-month results were similar (Table S4).

To investigate the potential influence of other factors that may influence dust concentrations over time, we estimated stratified models with a priori variables (Figures S2–S9) and evaluated if these findings were similar to our primary analysis. We did not see differences in the direction or magnitude of change in FR concentrations in our stratified models compared to results in the primary analysis (all homes), so these variables do not appear to solely explain the changes.

When we subset the data to homes that had furniture or foam removed with measurable FRs detected ≥1000 ppm, we observed substantial declines in PBDEs and TPHP concentrations (Figure 3). The chlorinated OPFRs (TCEP, TCIPP, and TDCIPP) had lower concentrations at each follow-up period after the furniture was replaced, although levels of TCIPP and TDCIPP slightly increased after an initial decrease at the six-month measurement. TPHP concentrations decreased and then remained at fairly constant concentrations. The small sample size limited our ability to do statistical testing with this subgroup of homes.

4. DISCUSSION

FR dust concentrations declined after older upholstered furniture or foam was removed from the home, with larger declines in the short-term compared to the longer-term. The clearest downward trends were observed for PBDEs and TPHP. The consistent decreases in PBDE concentrations after a major source was removed was not unexpected because PBDEs were phased out of use in 2004 and because PBDEs and their replacement, which includes TPHP, were the predominant FRs in TB117-compliant furniture. Concentrations of other chlorinated OPFRs did not decline as much as PBDEs in the longer-term measurements, likely because of their many other sources in addition to furniture foam, including textiles, plastics, adhesives, and rubber. The volatility of the OPFRs tend to be greater than PBDEs, which would result in greater anticipated declines in the home, given equal source removal, giving further strength to the idea that sources may remain.

Our results indicate that removing or replacing one of the largest sources of FRs in the home reduces concentrations of many FRs in the dust. However, we cannot separate the impact of the furniture replacement from the likely substantial cleaning event that may have taken place when the old furniture was removed. Results from an indoor model estimate that PBDEs have a residence time of several years with removal only through air exchange and typical cleaning rates, indicating that the greater reductions seen here result from actions taken (Shin et al. 2013). In other words, the large decrease in FR concentrations in most homes (particularly from pre-replacement to 6 months) may be partly due to removal of a large quantity of dust, an important reservoir of FRs, once furniture was removed. Our main model estimates stratified by HEPA vacuum use showed associations between using a HEPA vacuum and reductions in FR concentrations (Figure S2), and although FRs in the non-HEPA vacuum stratum also decrease, this may suggest the importance of cleaning on FR concentrations. However, the declines in FR concentrations cannot be fully explained by cleaning practices since we observed smaller reductions in phthalates between the pre-replacement and 6-month visit in the same study (data not shown). A study on the impact of intensive cleaning in the absence of furniture replacement could help determine other actions that effectively reduce FR exposures.

For many FRs, including BDE-99, BDE-100, TCIPP, and TDCIPP, the concentrations were lowest at six months and then increased at 12 and 18 months. The rebounding of concentrations may be due to those FRs re-equilibrating from other sources in the home after furniture or foam removal and the likely associated cleaning event, or introduction of other sources in the home that we did not capture. Despite the observed rebound, furniture or foam replacement did result in lower FR concentrations.

The pre-replacement geometric mean concentrations of the seven FRs in dust in our study were within the range of median FR concentrations reported in other studies of house dust in the US (Figure S11). We expect that changes in FR concentrations in other indoor environments replacing similar amounts of FR-furniture would be consistent with our findings.

While our study is the first we know of designed to evaluate the impact of TB117-2013 furniture replacement on FR concentrations in the home, there were several limitations. First, our study had a small sample size, which limited our ability to include variables that may have influenced the changes in FRs concentrations and the interaction between those variables in our main model. For example, HEPA vacuums tended to be used in larger homes, and we were not able to account for how those variables may interact with the changes in FR levels. Second, the post-replacement conditions, such as cleaning frequency, were not consistent across homes. While this variability represents realistic conditions, we were not able to account for all of these conditions due to our small sample size. Also, we were not able to standardize the foam and fabric collection component of the study; the voluntary replacement group participants volunteered collection of foam samples, while foam and fabric samples were collected from nearly all furniture items in the purchased replacement group because the study team was present when the older items were removed from the home. As a result, we were not able to identify FRs removed in all furniture or foam. In addition, there may have been FRs in furniture or other items introduced or not removed from the home that may have biased our results toward the null.

FR exposures and strategies to reduce exposures raise environmental justice concerns, even when those FRs have been phased out or banned. Biomonitoring studies have found associations between higher PBDE exposures and lower socioeconomic indicators (Adamkiewicz et al. 2011). Downstream effects of FRs in furniture have a disproportionate impact on socioeconomically disadvantaged communities (Betts 2015), perhaps through the second-hand market or waste disposal. Strategies to reduce FR exposures need to account for these exposure disparities. Our research evaluated an exposure reduction technique that incorporated replacing furniture foam, which may be a lower cost option, produces less waste by volume, and prevents moving furniture with FRs into the second-hand market. However, replacing furniture or foam both lead to FRs entering the waste stream. Innovation is needed to safely dispose and recycle discarded FR-furniture and FR-foam (Lucas et al. 2018), and we note that a life cycle analysis would have foreseen this end-of-life problem with FR-foam. We did not find substantial differences in FR reductions between homes that replaced their furniture foam versus those that removed or replaced entire pieces of furniture. Because replacing furniture foam is often more economical, this finding supports recommendations to replace foam cushions to reduce FR levels.

6. CONCLUSIONS

Because FR exposures are associated with many adverse health outcomes, reducing exposures is an important goal to improve public health. Furniture foam has been a major source of FRs in homes, and we found that replacing older upholstered furniture with FR-free or TB117-2013 furniture was an effective strategy to reduce FR exposures. Because replacing furniture items can be costly and can move FRs into the second-hand market, our findings also support replacing FR-treated furniture foam with FR-free foam as another way to reduce FR exposures.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Highlights

- TB117-2013 labeled furniture associated with lower flame retardant levels
- Flame retardant dust concentrations in house dust lower after older furniture replaced
- Flame retardant concentrations in dust remained lower a year after furniture replacement
- Flame retardant concentrations in dust decreased after replacement of furniture foam

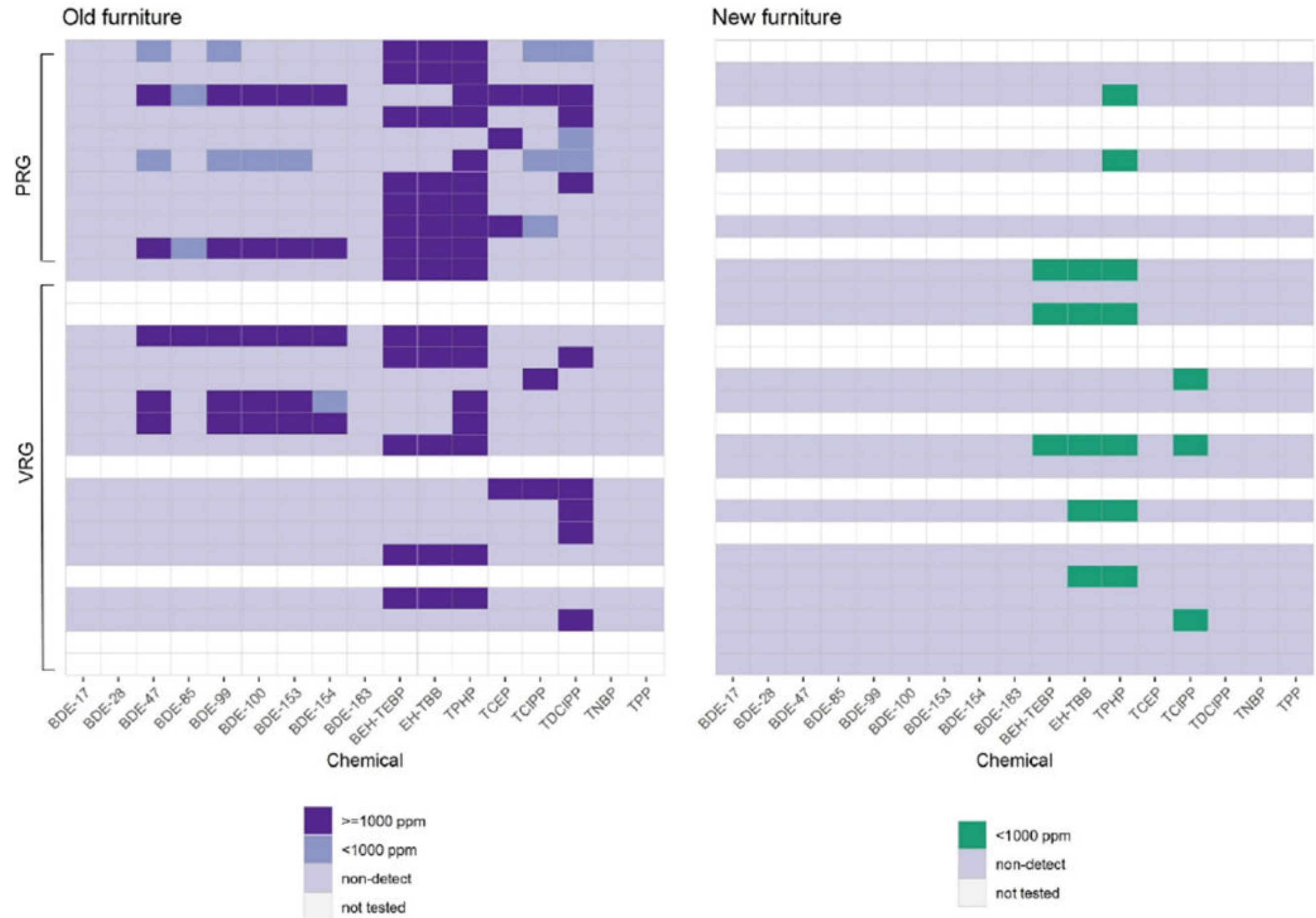

Figure 1.

Flame retardants detected above or below 1000 ppm in 23 homes with old furniture tested and 18 homes with new furniture tested. In some cases, multiple components of furniture, such as arm rest foam, cover fabric, decking material, and seating foam, were sampled. We used the highest value of all furniture samples in a given home to assign it as having a piece of furniture with flame retardants detected below or above 1000 ppm.

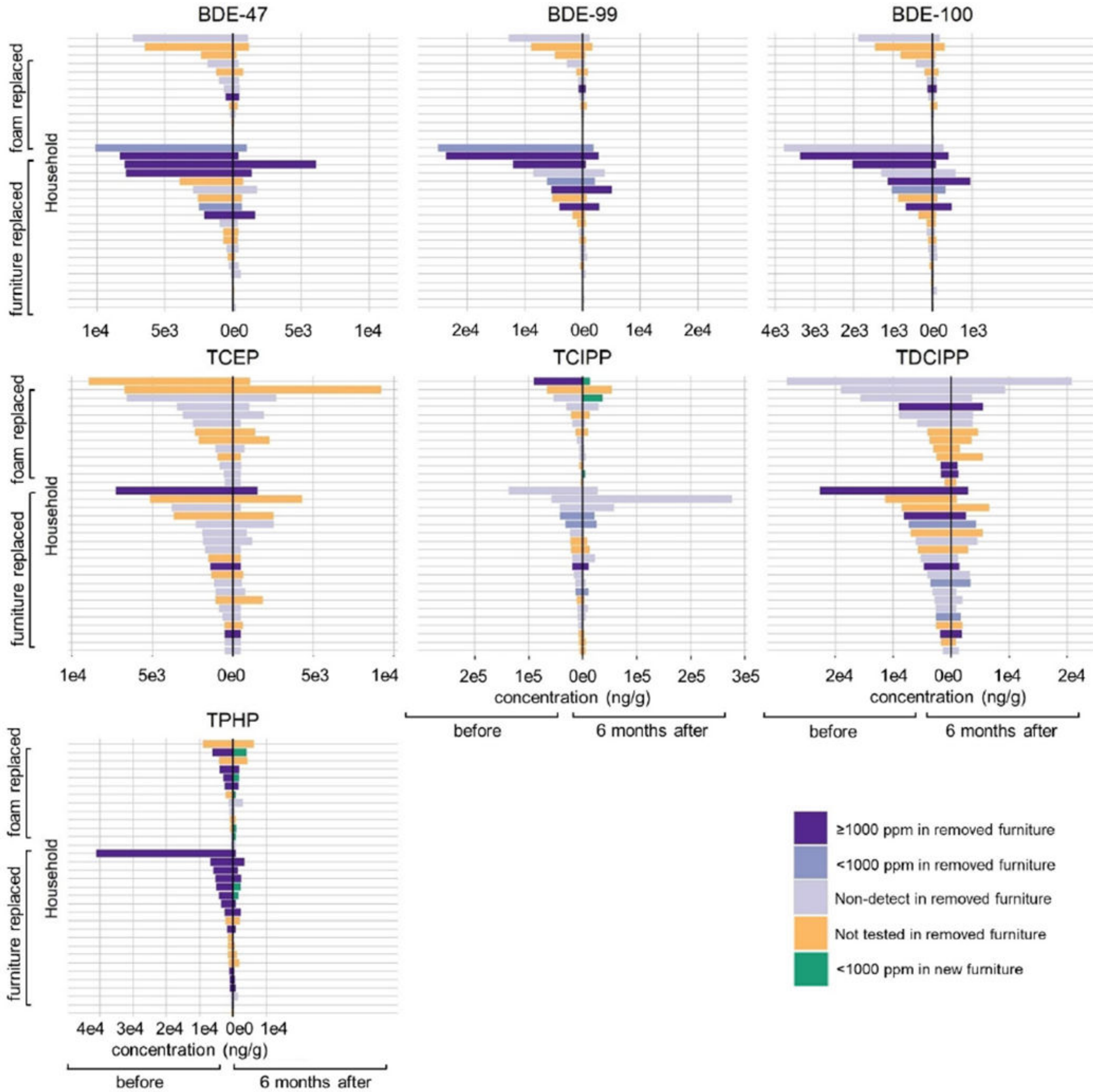

Figure 2.

FR concentrations in dust before and 6 months after furniture replacement. Concentrations are grouped by replacement type and sorted in descending order by group. Colors indicate the FR level detected in furniture foam or fabric.

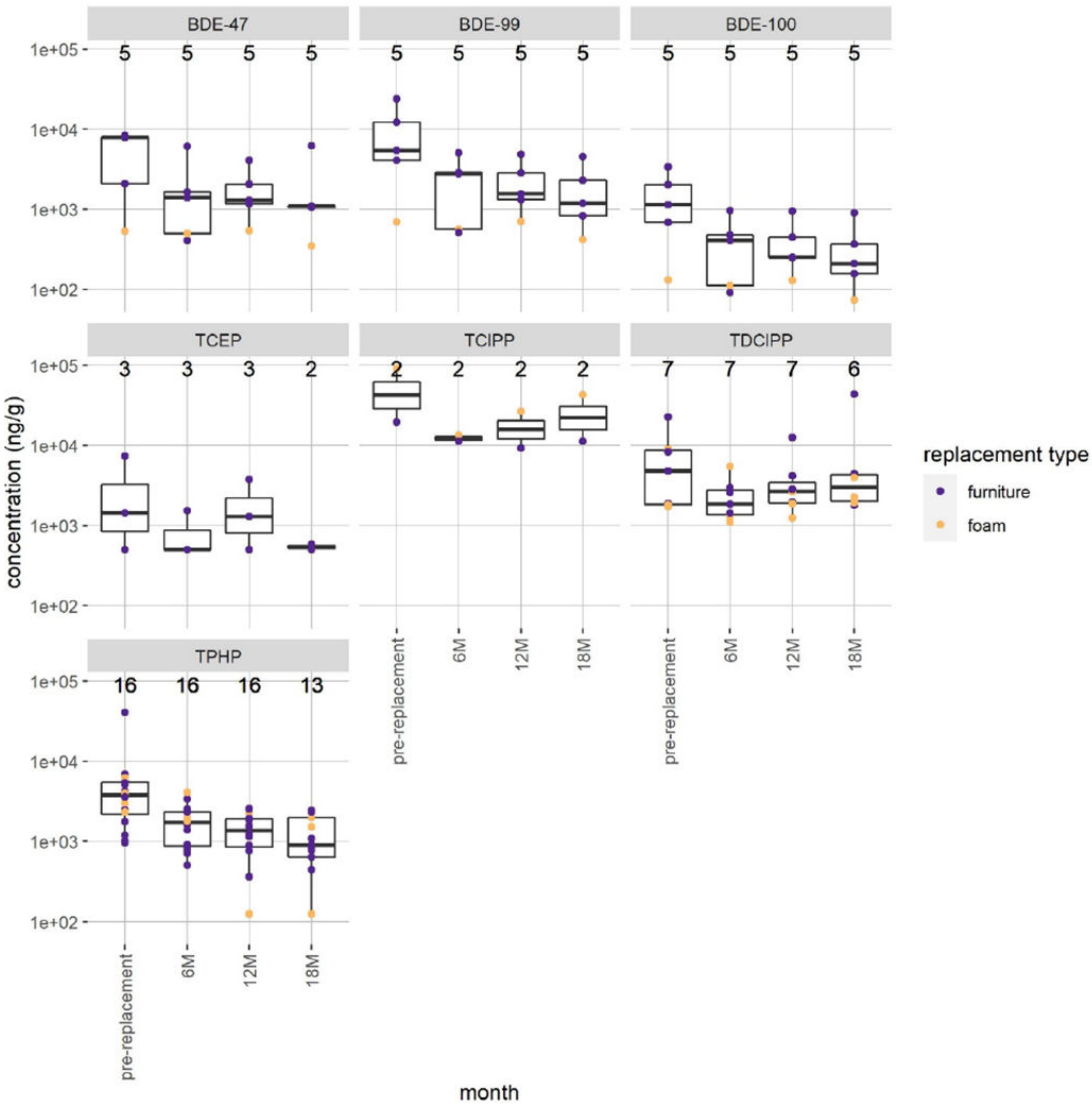

Figure 3.

FR concentrations in homes that removed furniture containing specific flame retardants before intervention and 6 and 18 months after the intervention. Number of homes sampled at each time point listed above boxplots.

Table 1.

Participation, furniture replacement type, and household characteristics for homes that completed at least one follow-up visit (n=33)

Replacement type	**Study visit follow-up**	**All**	**VRG**[1]	**PRG**[2]
Furniture				
	6 months	20	9	11
	12 months	20	9	11
	18 months	16	9	7
Foam				
	6 months	13	13	-
	12 months	12	12	-
	18 months	12	12	-
Participant characteristics		**All (n=33)**	**VRG (n=22)**	**PRG (n=ll)**
Income		n (%)	n (%)	n (%)
	<50,000	6 (18%)	0	6 (54%)
	50,000-100,000	4 (12%)	1 (5%)	3 (27%)
	100,000-150,000	10 (30%)	10 (45%)	0 (0%)
	>150,000	10 (30%)	10 (45%)	0 (0%)
	NA	3 (9%)	1 (5%)	2 (18%)
Education				
	GED or 12th grade completed	1 (3%)	0	1 (9%)
	1-3 years of college	7 (21%)	0	7 (64%)
	4 years of college	6 (18%)	3 (14%)	3 (27%)
	5+ years of college/graduate degree	19 (58%)	19 (86%)	0
Race/ethnicity				
	American Indian or Alaskan Native	0	0	0
	Asian	1 (3%)	0	1 (9%)
	Black or African American	3 (9%)	0	3 (27%)
	Hispanic	2 (6%)	0	2 (18%)
	Multi-racial	6 (18%)	5 (23%)	1 (9%)
	Native Hawaiian or other Pacific Islander	1 (3%)	0	1 (9%)
	White	19 (58%)	17 (77%)	2 (18%)
	don’ know/refused to answer	1 (3%)	0	1 (9%)
Vacuum with HEPA filter				
	Yes	21 (64%)	17 (77%)	4 (36%)
	No	8 (24%)	3 (14%)	5 (46%)
	N/A	4 (12%)	2 (9%)	2 (18%)
Vacuum frequency				
	< 1/week	10 (30%)	8 (36%)	2 (18%)
	≥ 1/week	23 (70%)	14 (64%)	9 (82%)
Main living area characteristics		**All (n=33)**	**VRG (n=22)**	**PRG (n=11)**
	Weighted % furniture replaced in living room	n (%)	n (%)	n (%)

Replacement type	Study visit follow-up	All	VRG[1]	PRG[2]
	≤ 75%	12 (36%)	10 (45%)	2 (18%)
	76-88%	4 (12%)	1 (5%)	3 (27%)
	89-100%	17 (52%)	11 (50%)	6 (55%)
Installed carpet				
	Yes	14 (42%)	3 (14%)	11 (100%)
	No	19 (58%)	19 (86%)	0 (0%)
Square foot		median (min-max) 206 (118-529)	median (min-max) 216 (118-529)	median (min-max) 153(123-294)

[1]VRG=voluntary replacement group

[2]PRG=purchased replacement group

Table 2.

Summary statistics for the FRs measured (ng/g) at four study visits

Abbreviation	Month	N	%<LOQ	median	mean	GM	P95	max	LOQ	LOD
BDE-47	Pre	39	87	950	2300	780	8000	10000	100	10
BDE-47	6M	33	91	430	710	380	1700	6100	100	10
BDE-47	12M	32	81	480	830	410	2600	4100	100	25
BDE-47	18M	28	86	370	1000	430	4400	6100	100	25
BDE-99	Pre	39	90	760	3800	1100	14000	25000	100	50
BDE-99	6M	33	91	530	940	520	3200	5000	100	50
BDE-99	12M	32	81	450	1200	530	3700	7000	100	50
BDE-99	18M	28	89	620	1500	570	5500	14000	100	50
BDE-100	Pre	39	64	160	600	190	2200	3800	100	10
BDE-100	6M	33	48	99 *	160 *	87 *	530	950	100	10
BDE-100	12M	32	50	99	200	110	670	1100	100	25
BDE-100	18M	28	54	120	250	110	880	2200	100	25
TCEP	Pre	39	69	1400	2400	1600	6800	8900	1000	500
TCEP	6M	33	39	630 *	1400 *	930 *	3300	9200	1000	500
TCEP	12M	32	47	880 *	1400 *	1100 *	3900	5500	1000	500
TCEP	18M	28	43	890 *	1200 *	980 *	3100	3400	1000	500
TCIPP	Pre	39	100	16000	25000	17000	69000	140000	100	100
TCIPP	6M	33	100	7500	21000	8400	55000	280000	100	100
TCIPP	12M	32	100	9300	23000	11000	68000	200000	500	500
TCIPP	18M	28	100	10000	17000	11000	47000	63000	500	500
TDCIPP	Pre	39	100	4100	6700	4600	22000	28000	100	100
TDCIPP	6M	33	100	2900	3500	2600	7600	21000	100	100
TDCIPP	12M	32	100	3300	4900	3700	12000	19000	100	100
TDCIPP	18M	28	100	4600	7200	4500	22000	43000	100	100
TPHP	Pre	39	100	1700	3500	2100	7100	41000	100	100
TPHP	6M	33	100	1300	1700	1300	4200	6400	100	100
TPHP	12M	32	75	890	1000	610	2400	2500	250	100
TPHP	18M	28	75	760	910	560	2400	2400	250	100

Pre=pre-replacement, GM=geometric mean, P95=95th percentile.

* indicates calculation is made when more than 50% of measurements were <LOQ.

Table 3.

Percent change in geometric mean (GM) dust concentrations estimated by linear mixed effects model between pre-replacement and 6 months and pre-replacement and 12 months for all participants and by replacement type.

		All participants	Furniture replacement	Foam replacement
Analyte	**Follow-up interval**	**Percent change in GM (95% CI)**	**Percent change in GM (95% CI)**	**Percent change in GM (95% CI)**
BDE-47	Pre vs. 6m	−48 (−64; −25)	−40 (−63; −1.4)	−56 (−75; −24)
	Pre vs. 12m	−45 (−62; −20)	−37 (−62; 3.1)	−52 (−73; −16)
BDE-99	Pre vs. 6m	−50 (−65; −28)	−45 (−66; −11)	−55 (−74; −21)
	Pre vs. 12m	−49 (−64; −26)	−46 (−66; −12)	−51 (−73; −13)
BDE-100	Pre vs. 6m	−52 (−66; −32)	−50 (−68; −22)	−52 (−73; −16)
	Pre vs. 12m	−41 (−58; −17)	−40 (−62; −6.8)	−39 (−65; 8.7)
TCEP	Pre vs. 6m	−43 (−55; −28)	−41 (−55; −22)	−46 (−63; −23)
	Pre vs. 12m	−31 (−45; −12)	−8.8 (−31; 20)	−56 (−70; −36)
TCIPP	Pre vs. 6m	−51 (−64; −33)	−48 (−64; −25)	−54 (−75; −17)
	Pre vs. 12m	−30 (−49; −4.9)	−23 (−47; 9.9)	−39 (−67; 12)
TDCIPP	Pre vs. 6m	−46 (−56; −32)	−53 (−64; −38)	−33 (−53; −4.9)
	Pre vs. 12m	−21 (−37; −1.8)	−19 (−38; 6.4)	−26 (−48; 6.1)
TPHP	Pre vs. 6m	−39 (−57; −13)	−52 (−70; −25)	−16 (−54; 54)
	Pre vs. 12m	−71 (−80; −58)	−65 (−78; −45)	−80 (−89; −62)

Pre-replacement to 6 month interval includes 33 participating households, Pre-replacement to 12 month interval includes 32 participating households, GM=geometric mean.