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Associations between flame retardant applications in furniture foam, house dust levels, and residents' serum levels

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ABSTRACT

Polyurethane foam (PUF) in upholstered furniture frequently is treated with flame retardant chemicals (FRs) to reduce its flammability and adhere to rigorous flammability standards. For decades, a commercial mixture of polybrominated diphenyl ethers (PBDEs) called PentaBDE was commonly applied to foam to fulfill these regulations; however, concerns over toxicity, bioaccumulation, and persistence led to a global phase-out in the mid-2000s. Although PentaBDE is still detected in older furniture, other FR compounds such as tris(1,3-dichloroisopropyl) phosphate (TDCIPP) and Firemaster® 550 (FM550) have been increasingly used as replacements. While biomonitoring studies suggest exposure is widespread, the primary sources of exposure are not clearly known. Here, we investigated the relationships between specific FR applications in furniture foam and human exposure. Paired samples of furniture foam, house dust and serum samples were collected from a cohort in North Carolina, USA and analyzed for FRs typically used in PUF. In general, the presence of a specific FR in the sofa of a home was associated with an increase in the concentration of that FR in house dust. For example, the presence of PentaBDE in sofas was associated with significantly higher levels of BDE-47, a major component of PentaBDE, in house dust ($10^{\beta} = 6.4$, $p < 0.001$). A similar association was observed with a component of FM550, 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), with levels that were approximately 3 times higher in house dust when FM550 was identified in the sofa foam ($p < 0.01$). These relationships were modified by dust loading rates in the living room and the ratio of sofa size to room size. Interestingly, levels of TDCIPP and tris(1-chloro-2-isopropyl) phosphate (TCIPP) were also higher in dust with detections in sofa foam; however, these associations were not statistically significant and may suggest there are other prominent sources of these compounds in the home. In addition, the presence of PentaBDE in sofa foam was associated with significantly higher levels of BDE-47 in serum ($p < 0.01$). These results suggest that FR applications in sofas are likely major sources of exposure to these compounds in the home.

1. Introduction

Flame retardants (FRs) are applied to polyurethane foam (PUF) in upholstered furniture to reduce its flammability. Implemented in 1975, the State of California's Technical Bulletin 117 (TB 117) mandated that all upholstered furniture pass a 12-s open flame test (State of California BEARHFTI, 2000). To pass these flammability tests, FR chemicals have been added to the PUF and sometimes other components of furniture such as textile coverings. The polybrominated diphenyl ether (PBDE) commercial mixture known as PentaBDE was a common FR used in furniture foam; however, it was phased out in the mid-2000s. In response, organophosphate FRs such as tris(1,3-dichloroisopropyl) phosphate (TDCIPP), tris(1-chloro-2-propyl)phosphate (TCIPP), and the commercial FR mixture Firemaster® 550 (FM550), which contains triphenyl phosphate (TPHP), isopropylated triaryl phosphates (ITPs), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB) and bis(2-ethylhexyl)-tetrabromophthalate (BEH-TEBP), were increasingly applied as replacements (Stapleton et al., 2012b). In a recent study analyzing over one thousand foam samples collected between 2014 and 2016, TDCIPP was found to be the most common FR detected in furniture foam (Cooper et al., 2016). Despite the phase-out of PentaBDE and the recent amendment to TB 117 in 2013, which replaced the open flame test with

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a smolder test, flame retardants, including PentaBDE, are still detected in home furnishings and other foam-containing consumer products. This is due, in part, to slow product turnover and recycling of products themselves as well as the use of PUF in applications (Cooper et al., 2016; State of California BEARHFTI, 2013).

PBDE exposure and toxicity have been studied and characterized over the last few decades. The PBDE congeners associated with the PentaBDE mixture have been measured extensively in the environment and in human tissues, and exposure has been associated with negative impacts on thyroid hormone regulation and neurodevelopment, both in human and animal studies (Costa et al., 2014; Hale et al., 2003; Herbstman et al., 2010; Hites, 2004; Rahman et al., 2001; Zhou et al., 2002). Although less is known about other FRs, research conducted in the late 1970s suggested that TDCIPP is mutagenic, and more recently, that it may also be neurotoxic (Babich, 2006; Behl et al., 2016; Dishaw et al., 2011; Freudenthal and Henrich, 2000; Gold et al., 1978). Additionally, TDCIPP was added to California's Proposition 65 List of Possible Carcinogens in 2011. In contrast to PBDEs, TDCIPP is rapidly metabolized and excreted. The metabolite, bis(1,3-dichloroisopropyl) phosphate (BDCIPP), has been measured ubiquitously in urine samples, suggesting widespread human exposure to TDCIPP in North America, Australia, and Europe; however, TDCIPP exposure appears to be higher in the U.S. population compared to other countries (Cequier et al., 2015b; Cooper et al., 2011; Dodson et al., 2014; Hoffman et al., 2017a; Van den Eede et al., 2015). Comparatively less is known about the toxicity of TCIPP, although it has been associated with endocrine disruption and limited neurodevelopmental changes in a few animal studies (Dishaw et al., 2014; Farhat et al., 2013). Like TDCIPP, exposure to TCIPP is also widespread. The hydroxylated metabolite of TCIPP has been ubiquitously detected in humans in several recent studies (Hammel et al., 2016; Van den Eede et al., 2013, 2015). Components of FM550 have been associated with endocrine and metabolic disruption in exposed rodents, and they have been shown to bind and activate nuclear receptors that regulate adipogenic pathways in in vitro models (Belcher et al., 2014; Fang et al., 2014; Patisaul et al., 2013; Pillai et al., 2014). Metabolites of both the organophosphate and brominated components of FM550, identified through dosed rodent studies and in vitro studies, are now frequently detected in human urine (Butt et al., 2014, 2016a; Hoffman et al., 2014; Phillips et al., 2016; Roberts et al., 2012). Notably, all of the aforementioned FR compounds have been widely detected in indoor house dust, which may serve as an important exposure pathway via hand-to-mouth activity (Bergh et al., 2011; Hoffman et al., 2015b; Stapleton et al., 2008; Van den Eede et al., 2012). In fact, several studies have found significant positive associations between PBDE levels in house dust and serum or breast milk in the US, reinforcing house dust as a primary exposure pathway (Johnson et al., 2010; Stapleton et al., 2012a; Watkins et al., 2012; Wu et al., 2007). More recent studies have suggested that diet may play a larger role than dust for exposure to PBDEs, especially in European populations where PentaBDE was not used as pervasively in furniture as in North America. One recent study found no association between PBDEs in dust and serum but did find significant associations with various dietary items (Cequier et al., 2015a).

Despite these studies, research linking specific FR applications in products to house dust or biomarker levels is limited. Previously, portable x-ray fluorescence (XRF) measurements of bromine in products found in the home were shown to be highly correlated with PentaBDE levels in paired house dust and serum samples (Allen et al., 2008; Imm et al., 2009). However, these portable XRF instruments are only sensitive to specific elements, such as bromine, and are not capable of differentiating between PBDEs or FM550, for example. Furthermore, XRF appears to have limited utility for chlorinated FRs (Stapleton et al., 2011). The presence of foam-containing napping equipment in early childhood education facilities also was associated with higher levels of tris(2-chloroethyl) phosphate (TCEP) and TDCIPP in dust (Bradman et al., 2014). A significant reduction in dust PBDE levels were observed with the removal of older furniture and carpets between sampling in 2006 and 2011; however, furniture was not verified to contain any specific FR chemicals, and the source of the reduction (e.g. furniture, dust-loading, or FRs in carpet padding) was not evaluated (Dodson et al., 2012). FR levels found on product surface wipes have been associated with dust levels; however, only prominent electronic products found in the home were evaluated, and most of the relationships were observed among these products and plastic casings, such as televisions and computers (Abbasi et al., 2016). In one study, counts of baby products used in the home were correlated with urinary BDCIPP levels in infants, highlighting a possible link between product use and exposure (Hoffman et al., 2015a).

In the present study, we sought to further examine specific relationships between FR application in furniture foam and human exposure. Our goal was to identify and quantify specific FR applications in PUF from study participants' sofas in the main living area and determine how the presence or absence of a specific FR related to levels measured in house dust and residents' serum levels. Further, we sought to determine whether characteristics of the furniture or living space modified this relationship. To our knowledge, this is the first study to compare levels of specific FRs in a consumer product, particularly levels in upholstered furniture, to a known biomarker and to house dust.

2. Materials and methods

2.1. Study design

Participants were recruited as a part of a case-control thyroid cancer study between April 2014 and January 2016. Papillary thyroid cancer patients treated at the Duke Cancer Institute and Duke University Medical Center were invited to participate in the study by their physicians, and willing participants were contacted by our study team for enrollment (n = 72). Control participants (n = 81) were recruited via flyers in the Duke University Medical Center facilities or randomly selected as other Duke patients undergoing routine wellness care or care for unrelated illnesses. We assume that case status does not affect concentrations of flame retardants in furniture, dust or serum; hence, we included both cases and controls in the current study. The study population is described in greater detail in Hoffman et al. (2017b). Once enrolled, study personnel visited each participant's home to conduct questionnaires and collect environmental samples and biospecimens. The participants in the study all lived within 50 miles of Duke University and had lived in their homes for at least two years, ensuring that their current homes reflected several years of past exposure. On average, participants in this study lived in their homes approximately 11 years. All study protocols and related materials were approved by the Duke Medicine Institutional Review Board for Clinical Investigations, and all participants gave informed consent before providing any information or samples.

2.2. Sample collection

All participants were instructed not to vacuum their homes in the two days before the visit. During each visit, the entire floor surface of main living area in the participant's home was vacuumed using a Eureka Mighty Mite vacuum fitted with a cellulose thimble in the hose attachment for dust collection (Stapleton et al., 2012a). The thimbles containing the dust were wrapped in aluminum foil and immediately frozen at − 20 °C. One small piece of foam (approximately 1–3 cm^3) was removed from the sofa located in the main living area, wrapped in foil, and immediately frozen. Almost all homes contained one sofa, but if other upholstered furniture (e.g. loveseats, chairs, ottomans, etc.) were present in the room, they were noted in the study log. Only PUF from the sofa, and not textile or fabric samples, were analyzed as part of this study. All participants provided non-fasting blood samples in serum-separator tubes which were centrifuged and frozen for analysis