



## Brominated flame retardants in offices in Michigan, U.S.A.

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### ABSTRACT

Brominated flame retardants (BFRs) are now ubiquitous contaminants with large reservoirs and high concentrations in buildings. Most of the information documenting BFR levels has been obtained in residences, and other environments that can lead to exposure have received relatively little attention, including offices that contain numerous BFR sources and where individuals spend considerable time. The aim of this study is to characterize BFR concentrations, potential emission sources, and migration pathways in office environments. We measure BFR levels in floor dust, indoor air, ventilation filter dust, and carpets in ten commercial and institutional buildings in Michigan, U.S.A. The median concentration of total BDEs in settled dust was 8754 ng g<sup>-1</sup>, at the upper range of levels previously reported. Especially elevated levels were found in offices in buildings that contained known or likely BFR sources, e.g., computer servers. A trends analysis in a newly constructed building showed remarkable increases in concentrations of BFRs in settled dust and indoor air, and apparent steady-state levels were reached 5 to 8 months after building completion, a particularly striking finding given that the building was constructed and furnished several years after the voluntary phase-out of the penta- and octa-mixtures. Airborne particulate matter collected in a building's HVAC system filters contained PBDEs, including BDE-209, at levels exceeding the concentration of floor dust. In conjunction with estimates of building air flow rates, filter efficiency and other parameters, mass balance calculations for this building were used to estimate the emission rates and reservoirs of PBDEs. The widespread distribution of BFRs found in offices in both new and old buildings suggests the significance of workplace exposures, the need for controls to minimize human exposure, intra-building migration, and environmental releases of these chemicals, and the need for monitoring in new buildings to confirm the effectiveness of the PBDE phase-out.

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### 1. Introduction

Recent years have seen disturbingly rapid increases in human exposures to brominated flame retardant (BFR) chemicals, including polybrominated diphenyl ethers (PBDEs) that are persistent, bioaccumulative and toxic (Johnson-Restrepo et al., 2005). These chemicals have been widely used in foams, textiles, plastics, electronic and other products since the 1950s (D'Silva 2004), and they are constituents of many materials and furnishings used in buildings. The high concentrations of BFRs found indoors relative to outdoor levels (Kemmllein et al., 2003) show that a fraction of BFRs in these materials and furnishings is released indoors, either as airborne vapor, particulate matter (PM), or incorporated into floor dust. BFR concentrations, emission sources and exposure pathways in buildings and other environments remain poorly understood, although exposure pathways likely include inhalation, ingestion and dermal contact (Sjödín et al., 2008).

Because indoor concentrations are so elevated, buildings are particularly important exposure sources, especially homes, workplaces and other "microenvironments" where people spend considerable time. Knowledge of concentrations in dust, air and food is critical information towards understanding these pathways and the culpable exposure sources.

A number of studies have reported that BFR concentrations are elevated in residences, including in the U.S.A. where only very recently have uses of these chemicals been restricted (Allen et al., 2008; Harrad et al., 2008; Batterman et al., 2009a). Information is more limited regarding levels in offices and other workplaces, although high concentrations have been reported in offices in Australia (Toms et al., 2009), the United Kingdom (Harrad et al., 2008), and Greece (Mandalakis et al., 2008). Data for offices in the U.S.A. are lacking, although the extensive use of BFRs in this country suggests that concentrations will be high.

The goals of this paper are to characterize BFR concentrations, potential emission sources, and migration pathways in office buildings. We report on concentrations of 21 BDE congeners and tetrabromobisphenol-a (TBBPa) in air and floor dust in offices in a diverse set of ten commercial and institutional buildings. The concentration data are compared to levels reported in the U.S.A. and elsewhere. In one of the buildings, newly constructed, we documented the temporal trend of BFR concentrations,

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examine carpets and ventilation filters as sources and reservoirs, and calculate a mass balance of BFRs among key building compartments.

## 2. Materials and methods

### 2.1. Building selection

Ten buildings containing offices that represented a cross-section of building types were selected using the following criteria: at least one area used as an office that had a suitable sampling location (sufficient space, electrical outlets, etc.); building access via permission from building owners; and convenience to the investigators. All of the buildings were in southeast Michigan, and most were in Ann Arbor, a city of 150,000 people located 60 km west of Detroit. Most of the buildings were identified from personal contacts of the study team, and several of the buildings contained areas used for non-office functions. The buildings varied greatly in many respects, ranging in age, ventilation system type, furnishings, and many other aspects. Building characteristics are summarized in Table 1, and additional details are given in Supplementary Materials A.

A preliminary visit investigated the suitability of each building. A walkthrough survey was then conducted to identify building characteristics, e.g., interior building and room volumes. We selected a central interior office area, as well as an outdoor location (roof or nearby site) for simultaneous monitoring over a one week period. Repeated measurements using identical protocols were made in four buildings (#1, 2, 7 and 9). A trend study was conducted at one of the buildings, newly completed, and included measurements of air, dust, ventilation system filters and carpets over a 15 month period using eight repeated measurements, starting prior to occupancy in summer 2006, and then every 2–3 months. At the time of the initial measurements, the building was fully furnished, but absent were paper files, books, personal belongings, etc.

### 2.2. Sample collection and analysis

Dust samples were collected from a central building area that did not have excessive occupant traffic. After marking off a 1 m<sup>2</sup> area, the area was vacuumed using an Oreck® XL compact canister vacuum and repeated strokes that reached across the sampling area. Occupants

were asked not to clean the room a week before sampling. If the building was resampled, the same area was sampled. Dust was collected on a pre-cleaned and pre-weighed filter placed immediately behind the vacuum's nozzle. Prior to use, filters were cleaned in Soxhlet apparatus using a hexane–dichloromethane mixture, dried, conditioned (25 °C, 33% relative humidity), weighed and stored individually. Prior to each use, the vacuum's hose and nozzle assemblies were cleaned with solvents, water, a soap solution, air-dried, and stored individually. The weight of the collected dust was determined gravimetrically after conditioning the filter at 33% relative humidity and 25 °C.

Airborne particulate and vapor samples were collected using medium-flow sampling systems, 47 mm dia 1 µm pore size PTFE filters (SKC Inc., Eighty Four, PA) housed in PTFE cassettes, which were followed by pre-cleaned polyurethane foam (PUF) elements (22×76 mm, SKC, Inc, Eighty Four, PA) enclosed in custom glass cartridges (Batterman et al., 2009a). The flow rate was 15 l min<sup>-1</sup> and a volume of 151 m<sup>3</sup> was collected over the one week sample period. Both PUFs and PTFE filters were stored in cleaned foil and polyethylene bags. When sampling outdoors, the filter cassette and PUF cartridge were placed in a weatherproof metal enclosure with the inlet located at approximately 1.5 m above the ground and at least 1 m from the nearest vertical surface.

In one building (#7), we analyzed samples from both new (unused) and exposed filters from the building's heating, ventilating and air conditioning (HVAC) system, as well as carpet. The HVAC system utilized 30% efficiency folded pleat panel filters (12×24×1", HI-E40D, Purolator, Henderson, NC), followed by 65% efficiency bag filters (12×24×22", PUR-FLOW-B PFB12-9655-Cust, National Air Filter, Carlstadt, NJ). Composite samples of panel and bag filters, totaling 100 cm<sup>2</sup> in area, were obtained by cutting five strips (2×10 cm) from different sections of randomly selected filters. The area and weight (after conditioning at 25 C and 35% relative humidity) were determined. These samples are referred to as "bulk filter samples." In our initial analyses, extraction efficiencies of the panel filters were acceptable, but the bag filters showed low and inconsistent recoveries, which appeared to result from the complex matrix, which included the multilayer filter (fabricated from both woven and non-woven layers, binders, stitching, and other materials),

**Table 1**  
Summary of buildings and sampling sites.

ID	Building name	Site description	Date(1)	Office location
1a	University Building 1	Classroom	6/5/06	Workspace: classroom M1138 (desks, chairs, podium, projector, projector stand)
1b	Same	Same	5/17/06	Same
2a	Equipment Sales/Service	Machine testing and distributing	6/20/06	Machine testing office (desks, file cabinets, machines)
2b	Same	Same	10/4/06	Same
3	Medical Equipment Manufacturer	Manufacturer of plastic products used for athletic protection and disability	7/5/06	Conference room and office.
4	Medical Equipment Manufacturer	Moderate-sized plastics plant, producing athletic protective gear and for disabled persons.	7/13/06	Clerical/office/reception area at far south end of complex
5	Art Museum 1	Art Museum and Education Department Office	7/26/06	Education department office: book shelf, 2 desks, 3 chairs, window along length of one wall, carpeted with overlying rug
6	Art Museum 2	Library and Periodical Room	8/7/06	Workspace: library and book stacks
7a	University Building 2	Faculty Offices and Staff and Student Offices	8/23/06	Faculty and staff offices, student cubicles, conference rooms (east tower).
7b	Same	Same	10/26/06	Same
7c	Same	Same	1/5/07	Same
7d	Same	Same	2/23/07	Same
7e	Same	Same	5/14/07	Same
7f	Same	Same	7/17/07	Same
7g	Same	Same	8/8/07	Same
7h	Same	Same	11/13/07	Same
8	Tire Store	Tire store/auto service	9/21/06	Reception, showroom, customer service, waiting area
9a	Computer Server Building 1	Computer Server Room and Office	11/7/06	Office with desks, file cabinets, book shelves, microwave
9b	Same	Same	12/4/06	Same
10	Computer Server Building 2	Electrical Engineering and Computer Science	11/15/06	Office space

Notes: "ID" = building identification number.

as well as the collected dust. Additional steps to analyze the bag filters included separating the (dust-containing) “filter fabric” (a relatively fragile, white, polyester material) and the “backing layer” (a coarse non-woven polypropylene pink material); extracting and analyzing these components separately, and determining the total filter loading as the sum of BFRs on the filter fabric and backing layer. This achieved satisfactory separation and recovery as shown by the resolution of all critical congener pairs except for one congener, BDE-49, due to matrix effects associated with these filter samplers.

Samples of new carpet were obtained from the study area as small plugs (2.5×2.5 cm) from which we derived three sample types: “bulk” (entire carpet cross-section); “face layer” (top wearing layer); and “backing layer” (bottom adhesive-containing layer). These sections, which were thought to possibly utilize different BFR formulations and to have different potentials to generate (and capture) dust, were manually separated using cleaned tools. The same carpet was used in office areas throughout the building.

All samples (air, PM, dust, carpet, and filters) were returned to the laboratory in BFR-free containers for extraction and analysis following methods described elsewhere (Batterman et al., 2009b). PUFs and filters were separately extracted within 24 h of collection, and dust samples were extracted within 48 h. After sampling but prior to extraction, each sample was spiked with 1 µL of a surrogate solution containing 40 and 6 µg mL<sup>-1</sup> of PCB-65 and PCB-166, respectively. Each sample was Soxhlet-extracted for 24 h using hexane and dichloromethane (4:1). Eluents were concentrated to 10 ml using a rotary evaporator, and then eluted with ethyl acetate (7%) in hexane through a clean-up column packed with 4 g of 10% sodium carbonate in granular anhydrous sodium sulfate, 4 g of Florisil (100–200 mesh; Fisher Chemical), and 0.1 mL of 4.5 M sulfuric acid. Final extracts were concentrated to 1.0 ml under N<sub>2</sub> in a 30 °C water bath. Prior to GC/MS analysis, each sample was spiked with 15 µL of internal standard mixture containing PCB-30 and PCB-204 (25 and 1.5 µg mL<sup>-1</sup>, respectively).

HVAC filter samples (panel, bulk bag, and separate layers) were cut into small pieces (3×3 mm), spiked with a surrogate mixture of PCB-30 and 166, soaked in acetone (25 mL) overnight, and Soxhlet-extracted for 36 h in hexane/dichloromethane (4:1). Whole panel filters were similarly extracted. Extracts were reduced to 10 mL in a rotary evaporator. Carpet samples were cut into small pieces using pre-cleaned tools, spiked with surrogates, Soxhlet-extracted, cleaned up, and analyzed similar to HVAC filters. Surrogate recoveries exceeded 75%, indicating acceptable extraction efficiency.

Samples were analyzed for 21 BDE congeners and TBBPa using GC/MS (Agilent 6890/5973, Palo Alto, CA), negative chemical ionization mode, a DB-5 column (30 m, 0.25 mm id, 0.25 µm film thickness; J&W Scientific, Folsom, CA, USA), a 2 µl (splitless) injection, and two runs. Target ions and retention times are shown in Supplementary Materials B. The first run quantified TBBPa and 20 BDE congeners using an oven temperature starting at 80 °C, held for 2 min, ramped at 10 °C min<sup>-1</sup> to 300 °C, and held for 46 min. To avoid decomposition (Stapleton et al. 2005), BDE-209 was quantified in a second run which used a temperature program starting at 80 °C, held for 2 min, ramped at 50 °C min<sup>-1</sup> to 300 °C, and then held for 40 min. Calibration standards included TPPBa and BDE-17, 28, 75, 49, 71, 47, 66, 100, 99, 85, 154, 153, 138, 166, 183, 190, 203, 208, 207, 206 and 209 (Cambridge Isotope Laboratories, Inc., Andover, MA, USA). Standards were run for a wide range of concentrations (100 to 5000 ng mL<sup>-1</sup>) that encompassed the range expected in field samples.

### 2.3. Air exchange rates (AERs)

AERs in offices and flows between two building zones were determined using perfluorocarbon tracers (PFTs) and the constant injection method. In brief, different tracer gases were released continuously in each zone, and AERs and interzonal flows were derived from measured

concentrations assuming a first-order multi-compartmental conservation of mass approach and steady-state conditions. Octafluorotoluene (OFT) was released in offices using two emitters placed near ends of the zone (average emission rate = 6.0 mg h<sup>-1</sup>). Hexafluorobenzene (HFB) was released in a different zone of the building (emission rate = 13.8 mg h<sup>-1</sup>). Airborne PFT concentrations were measured at two or more office locations using passive volatile organic compound samplers, which were analyzed using an automated short-path thermal desorption/cryofocusing system (Model 2000, Scientific Instrument Services, Ringoes, NJ) and GC/MS (Agilent 6890/5973, Palo Alto, CA). This method provides low method detection limits (MDLs; <0.02 µg m<sup>-3</sup>). PFT concentrations measured at different locations within the offices were usually within 20%, indicating good mixing, thus concentrations were averaged to obtain a zone-wide value. AERs and air flows were calculated using the PFT emission rates, building and zone volumes, the average PFT concentrations, and a two-zone fully-mixed steady-state dilution model. This method and its performance are detailed elsewhere (Batterman et al., 2006; 2007).

### 2.4. Quality assurance

MDLs for BFR measurements were calculated as three times the standard deviation (SD) of multiple low concentration analyses ( $n = 7$ ), and also as the measured concentration that produced a peak area with a signal-to-noise (S/N) ratio of approximately three. Several sets of PBDE mixtures (containing all target PBDE congeners) were used for these analyses: mixtures in hexane representing the final extracts; and mixtures simulating the filter/PUF extract after evaporation, fractionation, etc. We used the higher MDL estimate, which was usually given by the S/N method. Estimated MDLs are presented in Supplementary materials B. Volume-specific MDLs were used in calculations.

For PBDEs, each sample batch included blanks, checks using standard reference dust (NIST 2585 “Organic Contaminants in House Dust”), and linearity, drift, spike, recovery and precision tests. Recoveries for the NIST dust were 83 ± 7% for BDE-209 and 92 to 118% for the other congeners. In addition, calibrations were checked with the Analytical Center “Typhoon” Laboratory in Obninsk, Russia. To check drift, standards were run after every 5 samples, and results were accepted only when standards varied by less than 10%. Surrogate recovery was always between 75 and 95%, and averaged 88 ± 18% for PCB-65 and 93 ± 11 for PCB-166. Subsequent to the analyses in the present paper, we have adopted labeled BDE-209 as a surrogate and, using otherwise identical protocols, obtained average recoveries of 91 ± 6%. Field blanks were obtained in each sampling event. For air samples, cleaned filters and PUFs were brought out to each sampling location and temporarily opened to simulate placement in the sampling apparatus. For dust, cleaned filters were temporarily placed on the nozzle-hose assembly. Blank levels on filters were below MDLs; blank levels on several of the initial PUFs contained moderate levels of PBDEs, but more extensive cleaning (using the procedure detailed) resolved this problem. Side-by-side tests showed sampling precisions within 30%. Break-through of vapor-phase BFRs was evaluated using back-up PUF adsorbents and, in the worst case, breakthrough was <5%. A full performance evaluation of the sampling system is presented elsewhere (Batterman et al., 2009b).

For AER determinations, all samples were collected in duplicate along with blanks. Duplicate variation for the PFTs measurements was typically below 15%. At least two sampling sites were used in each office zone to confirm that the air was well-mixed.

## 3. Results and discussion

### 3.1. BFRs in settled dust

BFR concentrations in floor dust are summarized in Table 2. TBBPa and all target BDE congeners were detected in most samples. Due to the systematic changes in concentrations at the newly commissioned building (#7, discussed later), only the last



measurements at this building are included in this table. The median  $\Sigma$ BDE concentration in floor dust across the ten offices was  $8754 \text{ ng g}^{-1}$  (range from 1340 to  $38,917 \text{ ng g}^{-1}$ ), and the highest concentration congeners were BDE-209, 99, 47, 183 and 208 (based on medians). BFR concentrations across the buildings spanned a considerable range, as shown in Fig. 1. Across buildings (and after averaging replicates and repeated measurements), concentrations typically varied about two orders of magnitude; less variability was seen for BDEs-47, 85, 99, 100, 153, 154 and 209. The office in building 4, the medical equipment manufacturer, had the highest  $\Sigma$ BDE concentration ( $38,900 \text{ ng g}^{-1}$ ), followed by buildings 9 and 10, which contained spaces devoted to computer servers, with  $\Sigma$ BDE concentrations of  $33,700$  and  $24,300 \text{ ng g}^{-1}$ , respectively. The widespread use of octa- and deca-BDE formulations in electronic products, and migration of contaminants from the server areas may have been responsible for these concentrations. In building 10, an anomalously large peak eluted with BDE-190 (estimated as  $203,000 \text{ ng g}^{-1}$  equivalent of BDE-190), and was not considered to be a valid measurement. This peak likely was bis-tribromophenoxyethane (BTBPE), a relatively recent BFR sold as Solaris FR 20-1 for applications in plastics, elastomers, coatings and textiles. In future studies, we plan to characterize other non-PBDE BFRs, including BTBPE, using GC/MS in electron impact ionization mode and new external standards.

The repeated visits to buildings 2, 7 and 9 showed reasonably good reproducibility, generally within a factor of 5 to 10 for individual congeners. Only the latter visits to building 7 were considered representative due to the increasing trend seen in this building, as discussed later. Some of the poorer reproducibility seen at building 9, especially for the highly brominated PBDEs, may be due to the small amount of dust collected, which can impede accurate quantification.

Across the buildings, homologue and near-homologue groupings of BDEs were highly correlated in the dust samples, e.g., BDEs-47, 99 and 100 had Spearman rank correlation coefficients from 0.70 to 0.96, and BDEs-206, 207, 208 and 209 had correlation coefficients from 0.73 to 0.98. In contrast, concentrations of the BDEs in the penta-formulation had low correlation with the octa- and deca-formulation. A factor analysis using logarithms of concentration and varimax rotations identified four groups: (1) BDEs-207, 206, 209, 208, 138, 203, 166 and 75, representing the octa- and deca-formulation; (2) BDEs-154, 100, 99 and 47 (along with TBBPa), representing the penta-formulation; (3) BDEs-49, 66, 190, 28, 17, 85 and 71; and (4) BDEs-183 and 153 (Spearman correlation coefficient = 0.46). These groupings are linked to the major technical mixtures and indicate co-releases of BFRs from the mixture. We are unsure why TBBPa was linked with the four penta-BDEs in group 2; this may represent co-releases or co-releases.

Three studies have reported BFR levels in floor dust from commercial and institutional buildings. A Japanese study of 14 offices reported the lowest levels, e.g., the median concentration of BDE-209 was  $1110 \text{ ng g}^{-1}$  (range from 150 to  $70,000 \text{ ng g}^{-1}$ ; Suzuki et al., 2006). Three offices examined in Australia had  $\Sigma$  BDE concentrations from  $583$  to  $3070 \text{ ng g}^{-1}$  (higher than levels in five Australian residences in the same study; Toms et al., 2009). A Birmingham, U.K. study of 18 offices showed a median  $\Sigma$  BDE level of  $7400 \text{ ng g}^{-1}$  (range from 790 to  $280,000 \text{ ng g}^{-1}$ ; Harrad et al., 2008). BDE-209 was by far the dominant BFR in each of these studies. We found a similar range but somewhat higher concentrations, and again BDE-209 was the dominant congener, although high levels of the penta-formulation were also found. Also, BFR levels in office dusts are comparable to levels in house dusts. For comparison: Stapleton et al. (2005) reported a median  $\Sigma$  BDE concentration of  $4,250 \text{ ng g}^{-1}$  (range from  $780$ – $30,100 \text{ ng g}^{-1}$ ) in 17 homes, all but one in the Washington, D.C. area; Schecter et al. (2005) sampled vacuum cleaner dust in 8 homes in Dallas, TX and gave a median  $\Sigma$  BDE concentration of  $2,507 \text{ ng g}^{-1}$  (range from 705 to  $69,000 \text{ ng g}^{-1}$ ); Allen et al. (2008) examined 20 Boston, MA area homes and found a geometric mean  $\Sigma$  BDE concentration of  $13,723 \text{ ng g}^{-1}$  (range from 3020 to  $192,100 \text{ ng g}^{-1}$ ) in the main living area and considerably lower levels in bedrooms and vacuum cleaner dust; and recently we found a median concentration of  $21,000 \text{ ng g}^{-1}$  in southeast Michigan homes (maximum of  $290,000 \text{ ng g}^{-1}$ ; Batterman et

al., 2009a). The U.K. and the U.S.A. have much higher concentrations than other countries, reflecting usage patterns (Sjödin et al., 2008).

The specific sources and pathways that lead to high levels of BFRs in floor dust have not been determined. BFRs might be directly released as particles and fibers, or fragments thereof, which are shed from BFR-containing materials, e.g., foams in furniture and curtains, which are subsequently deposited as floor dust. A second pathway involves airborne BFR vapors, produced by volatilization or sublimation from aging BFR-containing materials, e.g., hot electronic components, which are then sorbed onto airborne particulate matter that is subsequently deposited as floor dust. A third possible pathway is that BFR vapors may be adsorbed directly onto floor dust. Airborne monitoring of BFRs (discussed later) does not clearly separate these pathways, even though we separated vapor and particulate fractions. An exploratory investigation using forensic microscopy found that bromine in house dust was concentrated on widely scattered and highly contaminated particles, suggesting the importance of abrasion or weathering of PBDE-containing materials, rather than volatilization, for samples with high concentrations of BDE-209 (Webster et al., 2009). However, sources and pathways may depend on the congener and material, and our data and the literature suggest considerable variability among buildings and the importance of partitioning among vapor and particulate phases.

We associated high levels of BFRs in floor dust in offices with several factors. First, the presence of many or strong BFR sources elsewhere in the building (not in the office being studied) elevated levels in offices, as demonstrated in the buildings containing computer servers (#9 and #10) that showed high concentrations of octa- and deca-BDE, and the buildings with foam processing operations (#3 and #4) that showed high levels of penta-BDE. Second, the ubiquitous nature of BFRs in all of the studied offices suggested the importance of the local sources, that is, in the offices themselves, as well as the general widespread use of the penta (and other) formulations in the U.S.A. Third, floor dust in offices, especially interior rooms, generally contains small loadings of sand, soil, grit and other materials that arise primarily from exterior soils and dusts, at least relative to the loadings in residences. Such coarse-size material is not expected to contain significant levels of BFRs, and also is unlikely to possess characteristics favorable for gas-to-particle sorption, e.g., a high surface area. High levels of grit and other coarse dusts will have the effect of “diluting” or lowering BFR concentrations (Harrad et al. 2008). Office floor dust tends to contain mostly smaller particles, fibers and less coarse grit, and a higher fraction may be derived from BFR-containing materials or with properties that are favorable to sorption. We did not characterize the type or origin of the dust, but the dilution effect is buttressed by a negative (though weak) correlation between dust loading and BFR concentrations (typical Spearman correlations from  $-0.1$  to  $-0.3$ ), and previous observations of generally low BFR concentrations in coarser dusts in residential garages (Batterman et al., 2009a), although we note that offices and garages clearly differ in terms of BFR sources, ventilation, and other regards. A fourth explanation might simply be the proximity of BFR sources in offices to the locations sampled.

### 3.2. Airborne BFRs

We detected 12 BDE congeners in the vapor phase (Table 2). Fig. 2 shows that tri-, tetra- and penta-BDEs (e.g., BDEs-71, 47, 99 and 100) were detected. The median  $\Sigma$  BDE concentration was  $1260 \text{ pg m}^{-3}$ . The office in building 4, a production facility for medical equipment, had the highest levels of most BDE congeners ( $\Sigma$  BDE concentration =  $17,200 \text{ pg m}^{-3}$ ); the other buildings had  $\Sigma$  BDE levels ranging from 21 to  $5700 \text{ pg m}^{-3}$ . For the predominant congeners in the air samples (BDE-47, 99, 100), concentrations in air and dust were strongly correlated (Pearson  $r$  between 0.59 and 0.92). Correlations for other congeners were weaker ( $<0.20$  for BDE-28 and 71), and  $\Sigma$  BDE had moderate correlation ( $r = 0.55$ ), reflecting contributions of octa- and deca-BDEs that were found only in dust. While the sample size is small, these results suggest that levels in dust and air

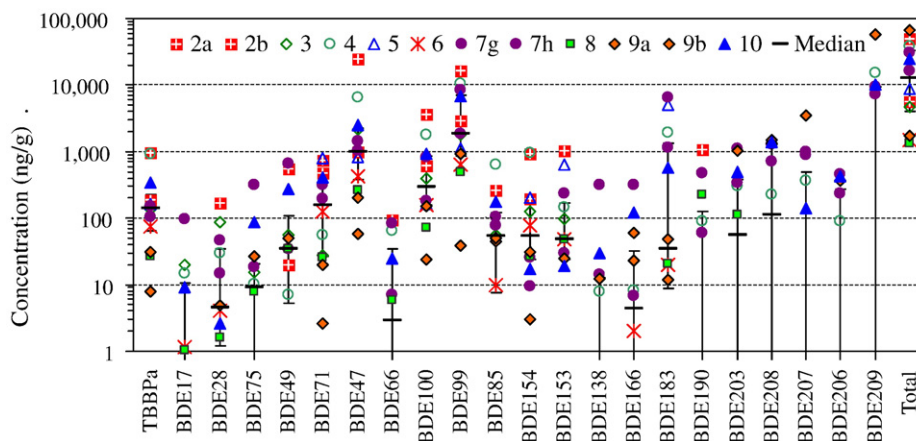


Fig. 1. BFR concentrations in dust in offices. Points show individual offices; median and 25th–75th percentile range also shown. Repeated samples to same building are shown for buildings 2, 7 and 9.

phases for at least the tetra- and penta-congeners are closely related and may in large part be explainable by vapor pressure and vapor–solid partitioning.

Airborne concentrations of BFRs in buildings have been measured in several studies, most of which have focused on residences. In homes in Ottawa, Ontario, the median vapor-phase  $\Sigma$ BDE concentration was  $100 \text{ pg m}^{-3}$  (Wilford et al., 2004). In Boston, MA,  $\Sigma$ BDE concentrations ranged from 220 to  $3500 \text{ pg m}^{-3}$  in living rooms and 170 to  $1500 \text{ pg m}^{-3}$  in bedrooms (Allen et al., 2008). Lower levels and different compositions have been seen in Europe (Harrad et al., 2006; Vorkamp et al., 2008). In Michigan homes, we recently reported a median  $\Sigma$ BDE concentration of  $3200 \text{ pg m}^{-3}$  and range from 340 to  $15,000 \text{ pg m}^{-3}$  using instrumentation and methods similar to that in the present paper (Batterman et al., 2009b). While our sample size is small, the levels in offices appeared to be comparable or perhaps somewhat higher than the other North American studies.

Results from PM sampling are also shown in Table 2. Because PM mass concentrations were low (median of  $8.8 \mu\text{g m}^{-3}$ ), only 1 to 2 mg of airborne PM was collected. Particulate-phase BDEs were detected in six of the ten offices. The sample from building 1 is anomalously high, and was collected during a period of sprinkler system testing. It is considered a statistical outlier and not discussed further. We detected congeners BDEs-47, 99 and 100, and  $\Sigma$ BDE concentrations ranged from 109 to  $774 \text{ pg m}^{-3}$ . The low mass of PM collected tends to inhibit detection of PBDEs, although concentrations on the order of  $1 \text{ pg m}^{-3}$  should have been detectable based on analytical sensitivities. We did not detect BDE-209 in the airborne PM, although later (Section 3.4) we show that BDE-209 was the most predominant congeners in airborne PM collected on ventilation system filters in building 7. Thus, we suspect our measurements of airborne particulate phase BFRs are underestimated.

Outdoor concentrations of BFRs in both vapor and PM at each building were much lower than indoor levels, and only a few of the particulate measurements exceeded MDLs. Thus, outdoor concentrations of BFRs provided only small or negligible contributions to indoor levels.

Most studies measuring airborne BFRs indoors have used passive sampling, which does not effectively capture the particulate fraction, and few if any studies have detected the more brominated BDEs in indoor PM, despite its strong partitioning. Our recent study of residences in Michigan also showed that BDEs-47 and 99 were the most prevalent congeners in airborne PM, and the median and maximum  $\Sigma$ BDE concentrations were 530 and  $3900 \text{ pg m}^{-3}$  (Batterman et al., 2009a). In the present study, offices showed similar patterns, but BDE (and PM) concentrations were somewhat lower. However, BFR levels across buildings showed great variability.

### 3.3. AERs and mixing in the offices

Air in most of the offices was well-mixed based on uniform PFT concentrations across the spaces (<15% variation), except buildings 6 and 8 on some visits when the variation approached 25 to 35%. Across offices, AERs averaged  $4.4 \text{ h}^{-1}$ , but spanned a large range ( $0.2$  to  $14.2 \text{ h}^{-1}$ ; Supplemental Materials C). Based on AER and ventilation systems, the offices fell into two groups. The first, including only mechanically-ventilated spaces within sealed buildings (buildings 1, 5, 7, 9), had high AERs (generally  $>3 \text{ h}^{-1}$ ) and high flows between zones. These buildings received a relatively large amount of ventilation air that was distributed (via fans and ducts) throughout the building with the potential to dilute indoor concentrations. The second group of buildings, including offices with operable windows or doors, had a mixture of natural and mechanical ventilation and lower AERs ( $<1 \text{ h}^{-1}$ ). Other things being equal, a lower AER permits the build-up of airborne concentrations due to emissions from indoor sources. While we did not find an association between AERs and BFR levels in air or dust, air flows measured between different zones of the building clearly showed the potential for contaminant migration to offices from strong PBDE sources in other parts of the building, e.g., the fabrication areas in buildings 2 and 3 where foam cutting/plastic molding operations took place, and the computer servers in building 9.

### 3.4. BFR trends and mass balances in a new building

We provided an in-depth assessment of BFR levels and trends in building #7 using eight sets of measurements conducted over a 15 month period. This sealed, mechanically-ventilated 7-story building has a floor area of  $8609 \text{ m}^2$ . It contains meeting rooms and a small cafeteria on the first floor; classrooms, meeting rooms and computer laboratories on the second; offices on the third, fourth and sixth floors; and laboratories on the fifth and seventh floors. A  $22.2 \text{ m}^3 \text{ s}^{-1}$  capacity variable air volume HVAC system served most of the building, including the office spaces studied on the 6th floor. Laboratory spaces were ventilated using a separate  $26.9 \text{ m}^3 \text{ s}^{-1}$  capacity HVAC system with 100% outside air (no recirculation). The study floor received approximately  $6.2 \text{ m}^3 \text{ s}^{-1}$  of ventilation air. BFR sampling was conducted in east and west office suites on the 6th floor. Both suites had perimeter offices and interior carrels made of metal, melamine, and fabric-covered surfaces. Floors had short-pile carpeting, and most offices and carrels had computers and other office equipment. The furnishings were new, however, many of the computers may have pre-dated the building by several years.

#### 3.4.1. Time trends

Prior to building opening in August 2006, BFRs in floor dust were at very low levels, e.g.,  $\Sigma$ BDE =  $145 \text{ ng g}^{-1}$ . Over the next eight months, each BDE congener and TBPPa increased by 100 to 1000 times, and  $\Sigma$ BDE levels as high as  $50,600 \text{ ng g}^{-1}$  were reached, although concentrations from 16,000 to  $31,000 \text{ ng g}^{-1}$  were more typical. Fig. 3A shows time trends and fitted exponential and sigmoidal models, which show a doubling time of 23 days and a levelling-off after about 6 to 8 months for  $\Sigma$ BDE levels in dust. The rapid increase and stabilization fit the sigmoidal model reasonably well ( $R^2 = 0.69$ ). The BFR composition remained similar over the study period, primarily reflecting the penta- and deca-formulations. All target BFRs were detected in dust, but BDE-209 was the predominant compound, increasing from  $82 \text{ ng g}^{-1}$  at the beginning of the study, to levels as high as  $48,000 \text{ ng g}^{-1}$  at mid-study, before stabilizing at  $6900$  to  $9300 \text{ ng g}^{-1}$  at the end of the study. TBPPa also increased markedly over the study period, from 0.4 to  $270 \pm 250 \text{ ng g}^{-1}$ .

Indoor air concentrations of BFRs also fit a sigmoidal trend, and a steady-state level was reached about five months after the building opened (Fig. 3B). Compared to dust, the rate of increase was slightly slower (doubling time of 31 days). Indoor air was dominated by tetra- and penta-congeners, mostly BDEs-47, 99 and 71, though tri-through hexa-BDEs were detected in most samples. Vapor-phase  $\Sigma$ BDE concentrations increased from about  $100 \text{ pg m}^{-3}$  at the beginning of the study to  $930 \text{ pg m}^{-3}$  at the end of the study.

This increase and apparent stabilization of BFR levels in both settled dust and airborne vapor are striking, novel, and disturbing results of this study. Despite the voluntary phase-out of PBDEs that started in 2004, these results suggest that there are significant sources of these PBDEs in products that remain in the market and that were used in this building, which was completed nearly two years later. Moreover, concentrations reached in the building are comparable or higher than levels reported in many earlier studies when PBDE use was not restricted. While building-to-building and study-to-study comparisons involve a number of caveats, especially since few measurements exist for office buildings, our results show a continuing need to identify and control PBDE sources, and to monitor levels in other new buildings to evaluate the effectiveness of the phase-out. At this point, we would expect that inventories of PBDE-containing materials would have been largely used-up, and PBDEs found in new office buildings might indicate a lack of compliance with the phase-out.

#### 3.4.2. HVAC samples

New panel (or prefilters) filters showed low levels of BFRs ( $\Sigma$ BDE =  $162 \text{ ng g}^{-1}$ ). This trace contamination may have occurred in storage or transit since BFRs were not used in their manufacture. After three months of use, the loaded filters showed very high BFR levels, e.g.,  $\Sigma$ BDE =  $10,600 \text{ ng g}^{-1}$  (for filter + dust) equivalent to a loading of  $1.37 \text{ mg m}^{-2}$  or a

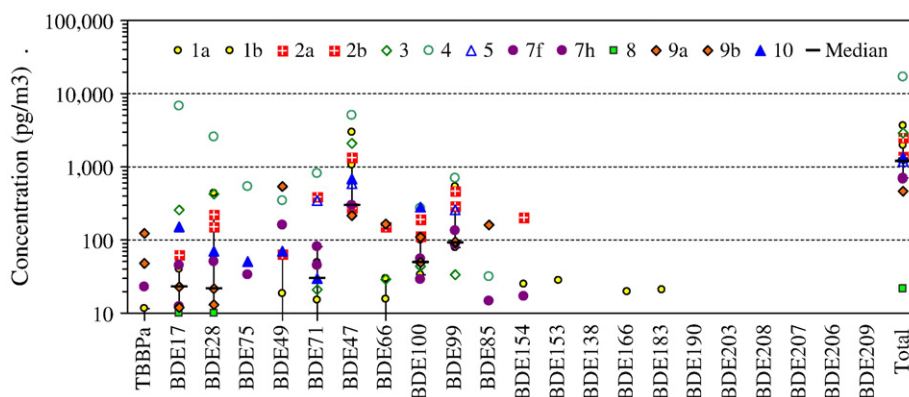


Fig. 2. Airborne vapor BFR concentrations in offices. Repeated measurements are shown for buildings 1, 2, 7 and 9. Otherwise as Fig. 1.

total of 39 mg collected in the filter bank (scaled up by filter area). New bag (or secondary) filters also showed only trace levels of BFRs ( $\Sigma\text{BDE} = 25 \text{ ng g}^{-1}$ ). After one year of use, concentrations were  $6600 \text{ ng g}^{-1}$ , equivalent to  $1.23 \text{ mg m}^{-2}$  or a total of 201 mg of BDEs in the filter bank. Interestingly, the panel and bag filters obtained similar loadings on an area basis, although their replacement schedule differed: panel filters are replaced quarterly, and bag filters annually. The much larger area of the bag filters accounts for the difference in the total accumulation in the filter bank. The HVAC system does not utilize fibrous insulation inside the duct work, although some (coated) sound proofing/insulation is present in the mixing and fan boxes. A visual inspection indicated that the HVAC filter dust appeared quite typical, and no fibrous insulation was seen.

Fig. 4 contrasts the BFR compositions on panel and bag filters. BDE-99, 85, and 153 (and coarse particles) were preferentially collected on panel filters; BDE-209 and octa- and nona-congeners (and fine particles) were preferentially collected on bag filters. The figure also shows the composition of settled dust, vapor and PM in the same building, using the average of the last three or four sampling events to increase representativeness. HVAC dust showed lower abundances of BDE-183 and other octa- and nona-congeners than settled dust, but overall compositions were fairly consistent. HVAC and settled dust could vary for several reasons; large dust particles are unlikely to be airborne; PBDE composition may vary by particle size distribution; and the HVAC system captures dust over large portions of the building while the floor dust samples are localized. Most importantly, analysis of the HVAC system filters indicates that octa-, nona- and deca-BDEs are present in indoor air. Detection limits and possibly other factors affecting air sampling performance permit the detection of only a small set of the more volatile PBDEs, at least when small to medium volume samples are collected.

### 3.4.3. Carpet samples

BFR concentrations in samples of new carpet were low ( $\Sigma\text{BDE} = 300 \text{ ng g}^{-1}$ ). This applied to face ( $\Sigma\text{BDE} = 690 \text{ ng g}^{-1}$ ) and backing ( $\Sigma\text{BDE} = 13 \text{ ng g}^{-1}$ ) layers. The face layer showed predominantly BDE-183 ( $400 \text{ ng g}^{-1}$ ) and BDE-209 ( $230 \text{ ng g}^{-1}$ ); levels of other congeners and TBPPa were below  $11 \text{ ng g}^{-1}$ . In comparison to dust, these concentrations are very low and may result from inadvertent contamination, although the congener patterns in carpet and dust differed (e.g., dust contained much higher abundances of TBPPa, BDE-47, BDE-99 and BDE-209, but lower abundances of BDE-193). The carpet did not utilize a sponge or foam rubber underlayment or cushion, which can contain BFRs. These results suggest that carpet was not a major source of BFRs in building #7.

### 3.4.4. Mass balance of BFRs

We derived a steady-state BFR and PM mass balance among major building compartments in building #7, depicted in Fig. 5. Parameters in these calculations were based on local conditions: HVAC filters were exposed to an average  $\text{PM}_{10}$  level of  $13 \mu\text{g m}^{-3}$ , a typical outdoor concentration; 30% outside air was assumed with the balance recirculated, with the HVAC duty cycle of 70% and flow rate of  $22 \text{ m}^3 \text{ s}^{-1}$ , taken from system specifications and typical operation; and panel and bag filter particle removal efficiencies were taken from Fisk et al. (2002). Based on these parameters, dust accumulations (loadings) were estimated as 600 g of  $\text{PM}_{10}$  in the panel filters over 3 months, and 4460 g  $\text{PM}_{10}$  in the bag filters over 1 y (the sampling periods). The 38 mg of  $\Sigma\text{BDE}$  determined for the panel filters is equivalent to a concentration of  $64 \mu\text{g g}^{-1}$  in the collected dust. For the bag filters (where 201 mg of  $\Sigma_{21}\text{BDE}$  was accumulated), the result is  $45 \mu\text{g g}^{-1}$ . These concentrations exceed measured levels in the building's floor dust by 1.5 to 2 times, which is reasonable since HVAC filter dust largely excludes sand, soil and other coarse and low sorption-capacity materials found in floor dust. We estimated that both HVAC filters would collect  $420 \text{ mg y}^{-1}$  of  $\Sigma\text{BDE}$ . To compare and confirm the BDE accumulations on HVAC filter dust, we calculated BDE fluxes through the filters using independent indoor and outdoor air concentrations measured in the building. With the measured indoor ( $930 \pm 290 \text{ pg m}^{-3}$ ) and outdoor ( $320 \pm 270 \text{ pg m}^{-3}$ ) concentrations, the  $\Sigma\text{BDE}$  concentration in the blended air passing through the HVAC system filters is  $750 \pm 220 \text{ pg m}^{-3}$ . (The outdoor samples were collected on the roof of the building, and concentrations were quite high compared to the literature, possibly reflecting entrainment of building air or local outdoor sources (especially since the building was just recently constructed). Since most air is recirculated, however, the mass balance results are not particularly sensitive to the outdoor air level.) With complete capture of vapor and particulate phase BDEs on HVAC filters,  $370 \text{ mg y}^{-1}$  of  $\Sigma\text{BDE}$  would be collected. This value is within 12% of that based on the HVAC filter measurements. This agreement is remarkable and implies that the HVAC filters capture most of the airborne BDE passing through the filters, including vapor and particulate phases.

For floor dust, we applied the measured  $\Sigma\text{BDE}$  concentration of  $33 \mu\text{g g}^{-1}$ , the measured dust loading of  $1 \text{ g m}^{-2}$ , and assumed that the dust loading reflected 2 months of accumulation. For the office area of  $3690 \text{ m}^2$ , the estimated  $\Sigma\text{BDE}$  generation rate is  $730 \text{ mg y}^{-1}$ . While this estimate depends on the dust loading rate, an uncertain quantity, it suggests that BFRs accumulate in floor dust at approximately twice the rate than in HVAC filters.

### 3.5. Study limitations

BFR concentration measurements in dust and vapor were generally reproducible and measurement precision was good, although uncertainties increased at low concentrations, and larger uncertainties were apparent in the outdoor air and PM samples. Sampling was conducted over one week periods, and included times when

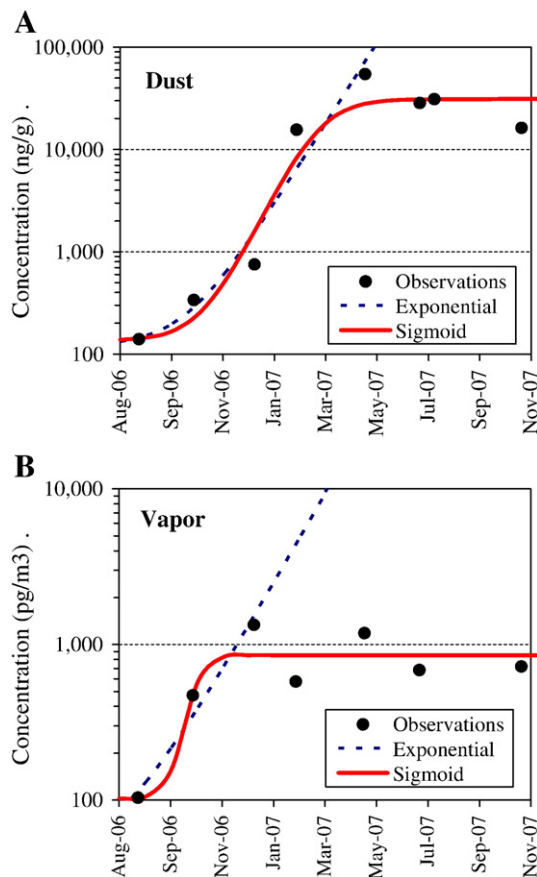
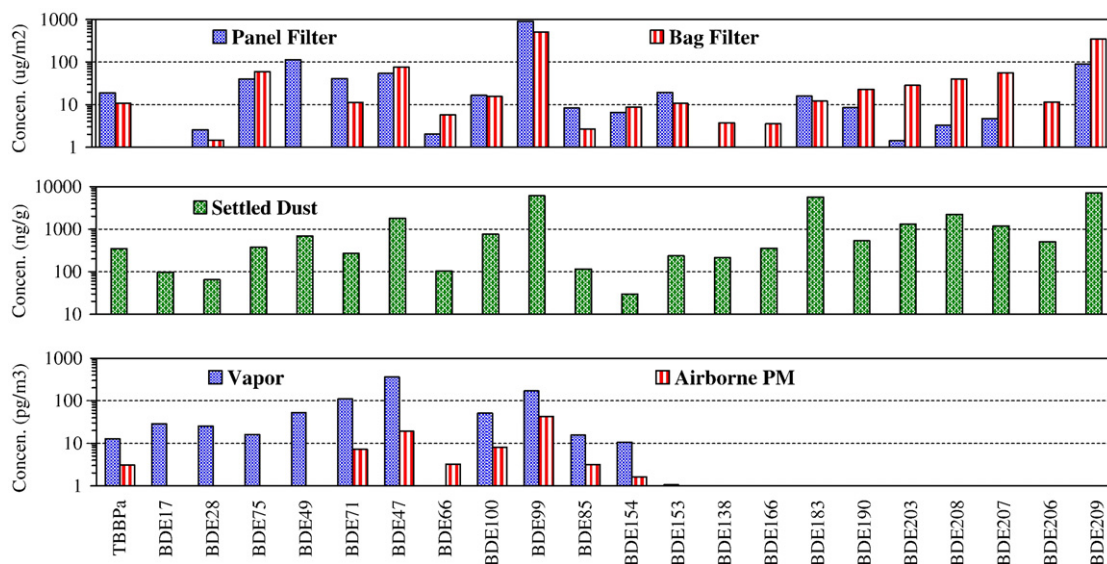


Fig. 3.  $\Sigma\text{BFR}$  levels in dust and airborne vapor in building 7, and fits to exponential and sigmoid models.

workers were not present. Across the study buildings, BFR concentrations varied considerably, as expected given the very different types of buildings and offices studied. The modest number of buildings tested, all drawn from a single geographic area, limits the study's generalizability, although the measured BFR concentrations are fairly similar to those reported in previous studies, and the commercial and institutional buildings in Michigan are expected to be typical of those in the North America. Within the buildings, BFR concentrations were seen to rapidly increase in the new building studied, but room-to-room variation and perhaps within-office variation warrants further investigation. Even newer buildings should be studied to document whether the PBDE phase-out is effective. In addition to BFR levels, other factors expected to demonstrate temporal and spatial heterogeneity include BFR emission rates, dust loadings and cleaning schedules. This suggests that sampling in additional building zones (and possibly using composite samples to reduce costs) would help to obtain samples that are more representative of the entire building. Finally, while based on best estimates of local conditions, the mass balance model required many assumptions, and further studies are needed in other buildings to confirm the results.

## 4. Conclusions

This paper presents some of the first measurements of BFR levels in U.S. offices, and several findings are striking. First, concentrations of PBDEs in settled dust and air are at the upper range of levels previously reported in general-use and residential buildings, probably due to the presence of many BFR-containing materials in offices, including computers, fabrics, and foam-containing furniture, and which constitute emission sources. Second, clearly elevated BFR levels were found in offices in buildings with known or likely BFR sources, e.g., computer servers and foam processing operations. The widespread distribution of BFRs in offices in both new and old buildings suggests that workplace exposures may be more important than previously recognized. Third, based on repeated measurements in a newly constructed building, concentrations of all measured PBDE congeners in settled dust and indoor air showed dramatic increases,



**Fig. 4.** Comparison of BFR composition in HVAC filters, settled dust, and airborne vapor and PM in building 7. Dust ( $n=4$ ), vapor ( $n=6$ ), and PM ( $n=6$ ) samples from May–Nov. 2007. Panel ( $n=2$  composite) and bag ( $n=2$  composite) filter samples (from HVAC system) are corrected for levels in new filters.

followed by an approach to steady-state levels that were reached 5 to 8 months after building commissioning. This finding is particularly remarkable given that the building was constructed and furnished after the voluntary phase-out in the U.S. of penta- and octa-mixtures. This indicates the need for further work to identify those materials used in new buildings that still contain PBDEs, and for additional studies in new buildings to gauge the effectiveness of the phase-out. The trends suggest that nearly a year is required to reach steady-state levels, and thus monitoring over a long time period is needed to evaluate effects of building interventions and other controls. Fourth, we showed that airborne particulate matter collected in a building's HVAC system contains high levels of PBDEs, including BDE-209, and in conjunction with estimates of building air flow rates, filter efficiency and other parameters, that mass balance calculations can be used to estimate the emission rates and reservoirs of PBDEs in the building. Sampling and analysis of HVAC filter dust may provide valuable information, although the sampling, extraction and analysis from these filters require special techniques. Finally, these findings suggest that HVAC filters, vacuum cleaner dust, and workplace zones handling BFR materials need special controls to minimize human

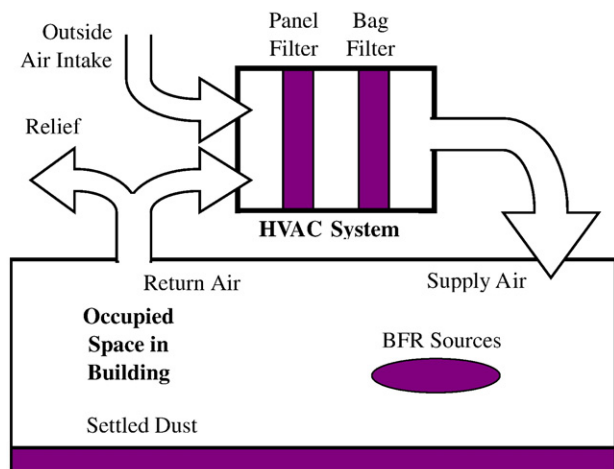
exposure, intra-building migration, and environmental releases of these materials from building sources.

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**Fig. 5.** Conceptual diagram of airflows and BFR reservoirs used in mass balance calculations of building 7.

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