Environmental Research 206 (2022) 112530

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Per-and polyfluoroalkyl substances (PFAS) and persistent chemical mixtures in dust from U.S. colleges

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ARTICLE INFO

Keywords:
Per- and poly-fluoroalkyl substances
Persistent organic pollutants
Polybrominated diphenyl ethers
Polychlorinated biphenyls
Carpets
Chemical mixtures

ABSTRACT

Indoor spaces contain several classes of persistent organic chemicals, including per- and polyfluoroalkyl substances (PFAS), polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs), and organochlorine pesticides (OCPs). However, concentrations of PFAS and persistent chemical mixtures and their associations with building characteristics on college campuses are understudied. We collected dust from 43 nonresidential spaces on four U.S. college campuses in 2016 and evaluated associations of room characteristics (carpeting, upholstered furniture, and years since last furnished) with dust concentrations of PFAS, PBDEs, PCBs, and OCPs. Nine PFAS, twelve PBDEs, two PCBs, and four OCPs were each detected in at least 75% of the spaces, including several chemicals (*e.g.*, DDT) that have been banned for decades. Concentrations were correlated within and, in some cases, between chemical classes. Wall-to-wall carpeting (compared to rooms without wall-to-wall carpeting) was associated with higher concentrations of six individual PFAS and a mixture of PFAS, and the number of pieces of upholstered furniture was associated with increased concentrations of a mixture of PBDEs. These findings indicate that carpeting and furniture are current sources of PFAS and PBDEs, respectively. Building and finish materials should be carefully selected to avoid exposure to persistent chemicals.

1. Introduction

Persistent organic chemicals are characterized by resistance to environmental degradation, long half-lives, and the potential to bioaccumulate (United Nations Environment Programme, 2017). Per- and polyfluoroalkyl substances (PFAS), polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs), and organochlorine pesticides (OCPs) are examples of chemical classes with these properties, each of which has been used widely in industry and consumer products (Glüge et al., 2020; California Environmental Protection Agency, 2019; Fernandez et al., 2021; U.S. Environmental Protection Agency, 2017a; b; U.S. Environmental Protection Agency, 2021a). Human exposure to these chemical classes is common in the U.S. (U.S. Centers for Disease Control and Prevention, 2019; U.S. Environmental Protection Agency, 2013), and previous research has identified a wide range of correlates of biomarker (Caspersen et al., 2016; Bradman et al., 2007; Sagiv et al., 2015; Horton et al., 2013) and dust concentrations for these compounds (Whitehead et al., 2014a; Rudel et al., 2008; Allen et al., 2008; Bradman et al., 2014; Rodgers et al., 2020). Indoor environments remain an important microenvironment for exposure to each of these persistent classes. Prior research has characterized concentrations of these chemical classes in dust in homes and in public spaces, including occupational settings (e.g., offices), fire stations, clothing stores, classrooms, and at childcare facilities (Zheng et al., 2020; Wu et al., 2020; D'Hollander et al., 2010; Harrad et al., 2019; Wu et al., 2019; Hall et al., 2020; Young et al., 2021a; Goosey and Harrad, 2011). However, few studies have examined characteristics of institutional spaces, such as universities, associated with dust concentrations for these chemicals. Further, to our knowledge, no prior study has identified correlates of mixtures of dust concentrations of PFAS, PBDEs, PCBs, and OCPs. Given the U.S. population is widely exposed to each of these persistent chemical classes, some of which have similar toxicological modes of action, identifying correlates of a mixture of PFAS, PBDEs, PCBs, and OCPs in dust can help identify intervention strategies to reduce exposures.

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https://doi.org/10.1016/j.envres.2021.112530
Received 9 September 2021; Received in revised form 19 November 2021; Accepted 4 December 2021
Available online 11 December 2021

PFAS are a class of over 9000 highly persistent chemicals (U.S. Environmental Protection Agency, 2021b) that are used for performance properties in consumer products, including carpets and furniture (Glüge et al., 2020; California Environmental Protection Agency, 2019). Their widespread use has led to ubiquitous human exposure (U.S. Centers for Disease Control and Prevention, 2019), where concentrations of various PFAS, including perfluorooctanesulfonic acid (PFOS), perfluorooctanoic acid (PFOA), and others, have been detected in homes, cars, childcare facilities, offices, universities, and other indoor environments (Wu et al., 2020; Haug et al., 2011; Fraser et al., 2013; Knobeloch et al., 2012; Kubwabo et al., 2005; Karásková et al., 2016; Gewurtz et al., 2009; Young et al., 2020). Studies that sampled dust in public spaces, even those that sampled dust more recently (e.g., 2014–2016) (Hall et al., 2020), consistently measured detectable levels of many PFAS, including those that have been phased-out, like PFOS and PFOA. These data underscore the persistence of PFAS in indoor spaces. Several studies also reported significantly higher PFAS in public spaces (e.g., fire stations, classrooms, offices) than in homes (D'Hollander et al., 2010; Harrad et al., 2019; Hall et al., 2020; Goosey and Harrad, 2011), suggesting public spaces are important indoor microenvironments for exposure to PFAS. Studies have found associations between PFAS levels in dust and serum and certain products, such as carpeting (Wu et al., 2020) and stain-resistant carpet or furniture (Boronow et al., 2019; Beesoon et al., 2012). Both PFOS and PFOA, along with other PFAS, continue to be detected in humans and in environmental samples years after their respective phase-outs (De Silva et al., 2021). However, hundreds of PFAS remain in active use in the U.S. and have failed to be systematically regulated by the U.S. Environmental Protection Agency (EPA) (U.S. EPA, 2017), resulting in the potential for continued human exposure for years to come.

Widespread exposure, even after phase-outs and regulatory action, is not limited to PFAS. PCBs (IARC, 2016), PBDEs (U.S. Environmental Protection Agency (EPA), 2016), and certain OCPs (U.S. Environmental Protection Agency, 2013) have been banned from production in the U.S.; however, these chemicals, some of which have been banned since the 1970s, continue to be found in indoor spaces (Rudel et al., 2003; Colt et al., 2005; Hwang et al., 2008; Whitehead et al., 2014b) and in human biomonitoring studies (U.S. Environmental Protection Agency, 2013; Whitehead et al., 2015; Bjorvang et al., 2021; Malliari and Kalantzi, 2017). Each of these classes have similarly been used widely in industry and in consumer products used indoors. PCBs, for example, were used in an array of indoor applications, such as electronics, paints, and adhesives (U.S. Environmental Protection Agency, 2021a). Contaminated dust is a major source of exposure for PBDEs, which were used primarily as flame retardants in furniture (Zota et al., 2008; Mitro et al., 2016). OCPs, such as chlordane and dichlorodiphenyltrichloroethane (DDT), were used indoors for termite and mosquito control, respectively, before they were banned in the U.S. (Eskenazi et al., 2009; ATSDR, 2018). Previous research has identified characteristics of indoor spaces associated with concentrations of chemicals in these classes. For example, presence of upholstered furniture, nap mats, and electronics have been associated with dust concentrations of PBDEs (Allen et al., 2008; Bradman et al., 2014; Rodgers et al., 2020), home age with PCBs and OCPs (Whitehead et al., 2014b), and flooring with PCBs (Rudel et al., 2008).

Persistent chemicals, including PFAS, but also PCBs, PBDEs, and certain OCPs, do not readily degrade, leading to continued increasing concentrations in the environment and therefore increasing possibility of known and not-yet-known health impacts (Cousins et al., 2019a). Breakdown of persistent chemicals may also lead to more toxic constituents; for example, relatively stable fluoropolymers can degrade to produce polyfluoroakyl acids (PFAAs), such as PFOA, which is linked to adverse health outcomes (ATSDR, 2021). As classes, PFAS, PBDEs, PCBs, and OCPs each contain chemicals or chemical groups listed on the Stockholm Convention for meeting criteria on persistence, bioaccumulation, long-range transport, and toxicity (United Nations Environment Programme, 2017). A uniform estimate of persistence across diverse chemical classes is difficult to obtain due to limitations in the applicability of current predictive models across a full range of chemical structures (Environment and Chemicals, 2017). Moreover, degradation of chemicals in environmental media such as soil or air may be different than in the indoor environment, given that degradation or transformation is slower indoors, and chemical fate indoors is largely determined by cleaning practices and air exchange (Shin et al., 2013). As a result, the introduction of persistent chemicals to indoor spaces is likely to have longer-lasting impacts than estimated by traditional degradation studies.

Exposure to each of these chemical classes has been linked with various human health outcomes. PFAS exposure, for example, has been associated with ulcerative colitis, kidney and testicular cancers, immunosuppression, and thyroid disease (Sunderland et al., 2019; Agency for Toxic Substances and Disease Registry (ATSDR), 2018). PCBs and PBDEs have both been linked with thyroid dysregulation (Allen et al., 2016; Pessah et al., 2019; Curtis et al., 2019), endocrine disruption (Rodgers et al., 2018; BuhaDjordjevic et al., 2020), and neurotoxicity (Pessah et al., 2019; Dorman et al., 2018). OCP exposures have been associated with cancer (Carcinogenicity of l, 2015), neurotoxicity (Briz et al., 2011), endocrine and immunological abnormalities (Mrema et al., 2013), and multi-generational risk of breast cancer (Cohn et al., 2015). Further, some chemicals in these classes have similar structures and biological activity, such as endocrine disruption (Allen et al., 2016; Rodgers et al., 2018; Kar et al., 2017; Bell, 2014; Mnif et al., 2011), and several studies have reported hormonal activity of indoor dust extracts containing several of these chemicals (Young et al., 2021b; Hamers et al., 2020; Kassotis et al., 2019). Their similar structures and biological mechanisms suggest that these chemicals may have additive or interactive health effects; for example, a mixture of PFAS was jointly associated with decreasing infant thyroid hormones levels (Preston et al., 2020), while a mixture of PFAS and PCBs was associated with decreased birth weight (Zhuang et al., 2021). These studies underscore the importance of identifying exposure to mixtures of these classes in indoor environments.

We sought to characterize the presence of these four classes as a mixture in indoor dust. Most exposure research has used a chemical-by-chemical approach, and we aimed to advance the current understanding of indoor exposures to these classes as a mixture. Several studies have used dimension-reduction techniques to classify mixture profiles within a chemical class in indoor spaces (Wu et al., 2010; Muenhor and Harrad, 2018; Zhang et al., 2011, 2016; Wang et al., 2013; Liu et al., 2011); however, we are not aware of any study that has simultaneously examined all four of these highly persistent classes in dust to identify correlates of exposure. We evaluated potentially important correlates of exposure with these chemicals on college campuses, because they tend to furnish spaces uniformly and use consistent furnishings within each space, which limits intra-space variability. These data can inform purchasing decisions at institutions like universities, as well as provide information relevant to regulatory actions on persistent chemicals that have the potential to affect exposures for years to come.

2. Methods

2.1. Sample collection

Trained staff collected 43 dust samples from nonresidential spaces on four New England college campuses in Spring 2016 (Rodgers et al., 2020). Nonresidential spaces included classrooms, dorm common areas, libraries, and lecture halls and auditoriums. Dust was collected using vacuum cleaners (Dyson, Inc., Chicago, IL) fitted with a custom aluminum crevice tool holding cellulose extraction thimbles (19 mm × 90 mm). In each space, staff lightly dragged the crevice tool over all room surfaces, including floors, furniture, desks, and window sills, for approximately 30 min. Samples were stored at < -20 °C prior to shipment to the analytical laboratory.

Information about the room characteristics was also collected concurrently with dust sampling. We examined several room characteristics as potential predictors of dust chemical concentrations. These predictors included the number of upholstered furniture (continuous), the number of years since the room was last furnished (continuous, obtained from campus administration or facilities personnel), and the carpet status of the room (yes/no, if the room had wall-to-wall carpet).

2.2. Chemical analysis

Prior to analysis, samples were sieved (<150 μm), weighed, and repackaged for shipment by Southwest Research Institute. One sample aliquot for each of the 43 field samples was analyzed at the University of Antwerp for five OCPs, five polybrominated biphenyls (PBBs), twelve PBDEs, and two PCBs. A second aliquot for each of the 39 field samples with sufficient remaining dust was analyzed at the U.S. EPA for ten PFAS. Full list of chemicals provided in Table S1.

Analysis of the OCPs, PBBs, PBDEs, and PCBs comprised two sample preparation methods with three extracts per sample. Briefly, samples were fractionated and either analyzed by gas chromatography–electron capture negative ion mass spectrometry (GC–ECNI/MS) and GC–electron impact (EI)/MS or by LC–MS/MS. Additional sample preparation and details on the instrumental analysis can be found in Rodgers et al. (2020).

Analysis of PFAS followed procedures outlined in Fraser et al. (2013), with minor alterations. In brief ~50 mg of sieved dust was sonic extracted with 5 mL of methanol spiked with 20 ng each of a suite of stable isotope labeled PFAS (Wellington labs) followed by centrifugation at 16,000 g. An aliquot of the supernatant (2 mL) was passed through a prewetted ENVI-Carb SPE cartridge (Supleco) and combined with 2 mM ammonium formate buffer (25:75 methanol:buffer) without evaporation for analysis. Extracted calibration curves from 1 to 100 ng (7 point) without any blank material added, as well as NIST Standard Reference Material (SRM) 2585 for analysis quality control, were prepared in the same manner. All samples were analyzed via isotope dilution MS/MS analysis on a Waters Acquity UPLC system coupled to a Quattro Premier XE mass spectrometer normalized by mass extracted and reported as ng PFAS/g dust.

2.3. Quality assurance/quality control

We used several quality assurance and quality control measures to evaluate the accuracy and reliability of our measurements, following approaches summarized in Udesky et al. (2019). We included three field blank samples. If a chemical was detected in all three blank samples, we blank-corrected by subtracting the median blank value from the reported concentrations; only PFHxS was blank-corrected, as all blanks for other compounds were non-detect (Table S2). For the PFAS data, the limit of quantification (LOQ) was set at 20 ng/g, and for the other compounds, the LOQs ranged between 1 and 5 ng/g dust and were calculated as three times the standard deviation of procedural blank values divided by the amount of dust used for analysis.

We evaluated accuracy using NIST SRM 2585 (Organic Contaminants in House Dust) for eight of the ten PFAS, PBDEs, two PCBs, and four of the five OCPs. The average relative difference between the certified or indicative values and the three SRM sample results was less than 20% for all PFAS except PFHxA, and less than 30% for the other persistent compounds, except for several BDE congeners (BDE 28, BDE 100, BDE 196, BDE 197, BDE 203) (Table S3). For measured standard concentrations of PFAS, average percent error ranged between 5 and 24%.

2.4. Data analysis

Values above the LOQ were considered “detects” and values below the LOQ, but still quantifiable, were considered “estimated values.” For samples reported as non-detect by the laboratory, we replaced values with the minimum value (including estimated values)*detection frequency. We calculated summary statistics and Spearman rank correlation estimates for chemicals with values by the laboratory in >75% of samples.

We used linear regression models to evaluate associations between room characteristics and PFAS concentrations. We modeled associations between room characteristics, including wall-to-wall carpeting (binary: 27 rooms with and 15 rooms without wall-to-wall carpeting), number of pieces of upholstered furniture (range: 3–725), year since the room was last furnished (range: 1972–2015), and natural log of dust concentrations of each chemical individually.

To investigate associations between room characteristics and a mixture of persistent chemicals, including PFAS, PBDEs, PCBs, and OCPs, in dust we used Principal Component Analysis (PCA). PCA was conducted with no constraints to assess the percent variation explained by the total number of principal components (PCs). We *a priori* chose to select the PCs that explained ~75% of the total variance in the data. Concentrations were natural log-transformed and z-standardized prior to PCA. We then used PC scores, which relate each observation in the dataset to the PCs using loading values, as the dependent variable in multivariable linear regression models. As with the individual chemical regression models, we included carpet status, number of upholstered furniture, and years since last furnished as covariates.

3. Results

3.1. Dust concentrations

PFAS were widely detected in dust. The percent of samples above the LOQ ranged from 23% to 87%, with the highest percentages for PFOA (87%), perfluorononanoic acid (PFNA) (85%) and perfluorohexanoic acid (PFHxA) (77%). All PFAS, except perfluorobutanesulfonic acid (PFBS), had values reported by the lab above 75%. PFOA had the highest geometric mean (GM) (96 ng/g), median (100 ng/g) and maximum (2400 ng/g), followed by PFNA (74 ng/g), and PFHxA (35 ng/g), and perfluorodecanoic acid (PFDA) (GM of 32 ng/g). Conversely, PFBA (9.6 ng/g), perfluoropentanoic acid (PFPeA) (11 ng/g), PFHxS (5.1 ng/g) had the lowest GMs (Table 1; Table S4).

The other persistent chemicals were also widely detected in dust. DDT, banned in the U.S. in 1972, was detected in 93% of dust samples. Two PCB congeners were detected in nearly all spaces. BDE 209, major congener in the DecaBDE flame retardant mixture, was found in all samples and at the highest concentrations of all targeted chemicals (GM of 8200 and maximum of 3,500,000 ng/g).

Concentrations in each sampling location were not uniformly high across chemical groups (Figure S1). However, within a chemical class, multiple individual chemicals were found at elevated concentrations (>75% percentile). Several sampling locations had elevated concentrations of individual PFAS and PBDEs.

3.2. Associations with room characteristics

Rooms with wall-to-wall carpet had higher concentrations of several PFAS compared to rooms without wall-to-wall carpet (Fig. 1), whereas the number of years since the room was last furnished and number of pieces of upholstered furniture was not associated with any PFAS (Table S5). Rooms with wall-to-wall carpet compared to rooms without wall-to-wall carpet had higher estimated marginal mean concentrations of PFPeA (15 ng/g versus 5.4 ng/g), PFHxA (56 ng/g versus 15 ng/g), PFHxS (8.5 versus 1.6 ng/g), perfluoroheptanoic acid (PFHpA) (28 ng/g versus 2.3 ng/g), PFOA (183 versus 38 ng/g), and PFNA (115 ng/g versus 32 ng/g), after controlling for the other room characteristics (Fig. 1). Concentrations of other PFAS were also higher in carpeted rooms, although not significantly.

Furniture was associated with concentrations of three PBDE

Table 1
Summary of dust concentrations (ng/g) from samples (n = 39 for PFAS, n = 43 for other chemicals) collected on college campuses.

Compound	LOQ[a]	% > LOQ	mean	std dev.	median	GM	95th %tile	max.
PFAS								
PFBA	20	26	14	11	<LOQ	9.6	35	48
PFPeA	20	23	21	27	<LOQ	11	83	130
PFHxA	20	77	79	130	38	35	210	800
PFHxS	20	26	36	95	<LOQ	5.1	150	540
PFHpA	20	49	90	210	<LOQ	13	270	1200
PFOA	20	87	270	490	100	96	1100	2400
PFOS	20	56	140	280	41	20	1000	1000
PFNA	20	85	170	360	79	74	340	2000
PFDA	20	62	180	440	53	32	750	2500
PBDEs								
BDE 28	1	81	37	79	13	11	140	480
BDE 47	1	100	2800	4700	870	820	14,000	20,000
BDE 85	2	100	240	540	57	60	860	3100
BDE 99	1	100	3300	5500	920	1100	16,000	26,000
BDE 100	1	100	790	1700	190	190	3500	8700
BDE 153	2	98	440	1100	83	99	1300	6900
BDE 154	2	100	400	810	120	110	2000	4500
BDE 183	2	91	38	60	13	19	170	250
BDE 196	2	98	270	870	26	42	760	5600
BDE 197	2	81	30	68	9.4	12	95	430
BDE 203	2	84	69	210	8.4	14	250	1300
BDE 209	5	100	150,000	570,000	5500	8200	900,000	3,500,000
PCBs								
CB 153	1	98	350	910	29	36	2000	4700
CB 180	1	98	400	1500	20	28	920	9300
OCPs								
CC	1	84	22	87	3.3	4.2	58	560
TC	1	91	29	110	4.1	5.3	100	700
TN	1	91	15	53	2.5	3.4	58	340
pp-DDT	2	93	50	61	24	26	200	210

[a] Limit of Quantitation (LOQ).

congeners. PBDE 100 and PBDE 153 concentrations were positively associated with years since last furnished and PBDE 209 concentrations were positively associated with number of pieces of upholstered furniture (Table S6).

3.3. Correlations within and between chemical classes

Concentrations of chemicals within the same class were the most correlated (Fig. 2). Spearman correlation estimates ranged from 0.23 to 0.98 for PBDEs, 0.66 to 0.95 for OCPs, and 0.13 to 0.91 for PFAS. Among the PFAS, sulfonic acids were more highly correlated with each other, as were carboxylic acids. We also found some moderate correlations across chemical classes, including DDT with some PBDEs (highest correlation of 0.56 for PBDE 196), PFOS with some PBDEs (highest correlation of 0.58 for PBDE 196), and PCBs with OCPs (highest correlation of 0.43 for PCB 153 and CC) and PFHxS (highest correlation of 0.47 for PCB 180).

3.4. Principal Component Analysis

We found that 4 PCs explained approximately 75% of the variance in the data: PC-1, 34%; PC-2, 18%; PC-3, 14%; and PC-4, 11%, respectively. Loadings are shown in Fig. 3, where the direction reflects positive and negative correlations of the chemical with the PCs. PC-1 was characterized as a mixture of all persistent chemicals and explained variance (loading >10%) for all chemicals across all classes, except PFHxS. PC-2 was characterized as a mixture of PFAS and lower brominated (<6 bromines) PBDEs. PC-3 was characterized as a mixture of PCBs, OCPs, lower brominated PBDEs (<6 bromines), higher brominated PBDEs (>8 bromines), and PFAS. PC-4 was characterized as a mixture of higher brominated PBDEs (>7 bromines), PCBs, PFBA, and PFOS. See Supporting Information for a full description of each PC loading.

In mutually adjusted models, rooms that had wall-to-wall carpet had significantly higher PC-2 scores ($\beta = 1.9$, 95% CI = 0.19, 3.6), indicating that highly carpeted rooms had higher concentrations of all PFAS compared to rooms without wall-to-wall carpeting (Table 2). An increase in the number of upholstered furniture was associated with decreased PC-3 scores ($\beta = -0.0046$, 95% CI = −0.0092, 0.000032) and increased PC-4 scores ($\beta = 0.0033$, 95% CI = −0.0002, 0.0068), both associations were marginally significant ($p = 0.051$ and $p = 0.063$, respectively). This indicates that an increase in the number of furniture is associated with increased concentrations of PCBs (PC-3), OCPs (PC-3), higher brominated PBDEs (PC-3, PC-4), and sulfonic acid PFAS (PC-3, PC-4). We did not find any associations between PC scores and year since last furnished.

4. Discussion

We detected multiple PFAS and other persistent organic chemicals on college campuses, many of which have been banned or phased-out for decades in the U.S. Concentrations were strongly correlated within each chemical class and, in some cases, moderately correlated between chemical classes. Presence of wall-to-wall carpeting was associated with higher concentrations of several individual PFAS and a mixture of PFAS.

Among the PFAS, we found the highest dust concentrations for PFDA and PFOA. Our results are generally consistent with a recent study of PFAS in dust on a college campus that found similar GM concentrations for PFOS, PFOA, PFHpA, PFPeA, PFNA, and PFHxS in rooms without renovations (Figure S2) (Young et al., 2020). PFOA concentrations were higher in our study (GM 96 ng/g vs. 11.6 ng/g), and we detected PFNA and PFDA more frequently (Young et al., 2020). However, PFHxA had the highest GM concentration (326 ng/g) in Young et al., which was nearly 10x higher than our study (35 ng/g). Similar to our study, PFOS and PFOA were found at the highest mean concentrations among PFAS in several U.S. household dust studies (Fraser et al., 2013; Knobeloch et al., 2012; Byrne et al., 2017; Strynar and Lindstrom, 2008). We found higher mean concentrations of PFDA, PFNA, and PFPeA compared to studies conducted in homes (Figure S2), but generally lower concentrations for PFHpA, PFHxA, PFOA, and PFOS (Fraser et al., 2013;

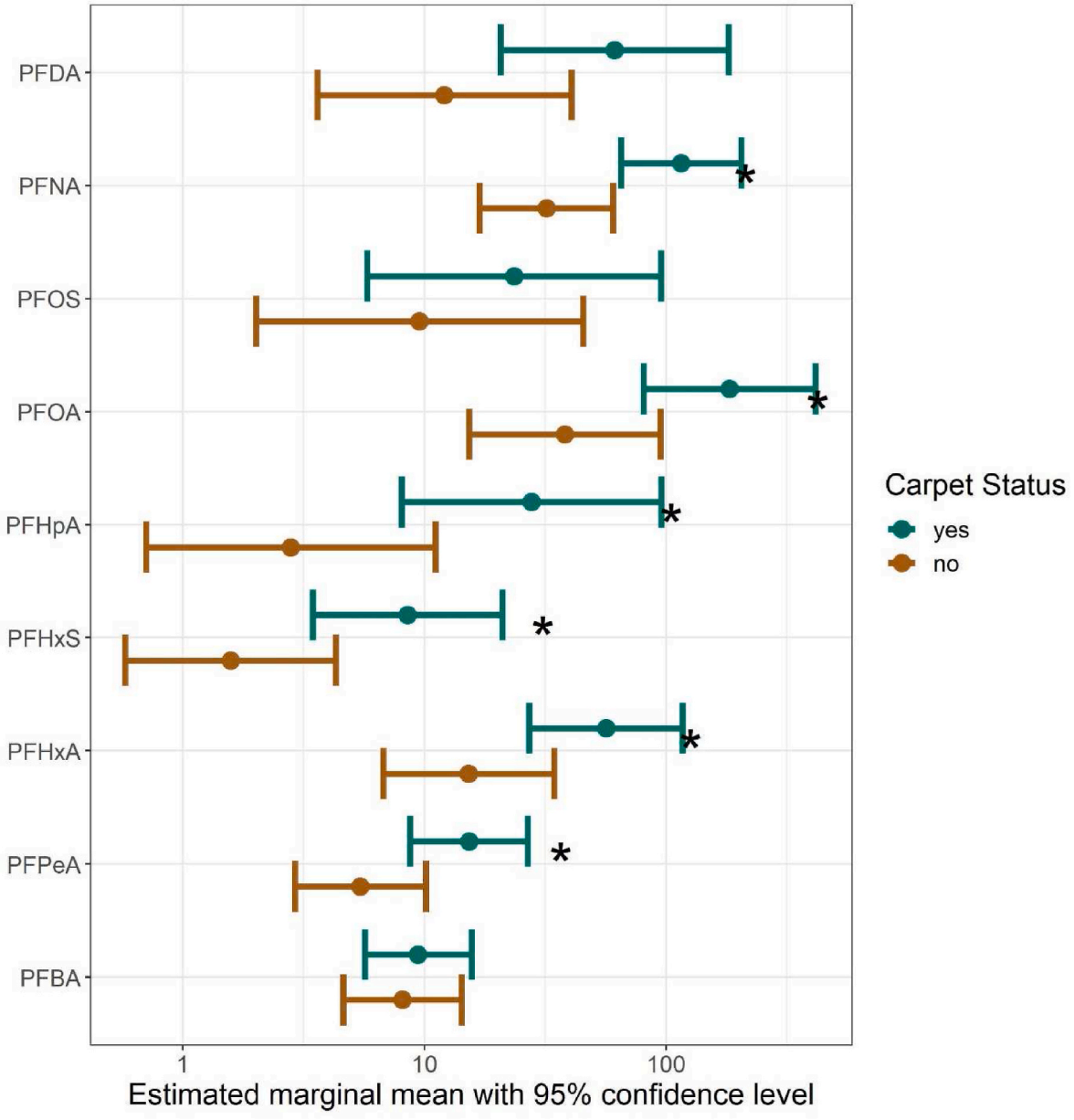

Fig. 1. Estimated marginal means (ng/g) with 95% confidence levels for rooms with and without wall-to-wall carpet, controlling for years since last furnished and number of upholstered furniture. Significant differences indicated by asterisk.

Knobeloch et al., 2012; Strynar and Lindstrom, 2008). Although this may reflect differences in when samples were collected, we found higher mean concentrations for most PFAS on college campuses compared to childcare centers in recently published studies in the U.S. (Zheng et al., 2020; Wu et al., 2020; Fraser et al., 2013)

Concentrations of several PFAS (PFPeA, PFHxA, PFHxS, PFHpA, PFOA, PFNA) were associated with carpet status, which is consistent with other studies (Haug et al., 2011; Gewurtz et al., 2009). Because only PFAS were associated with the presence of carpet, these findings suggest that carpets were current sources of PFAS. Our PCA confirmed this, where PC-2, comprised of PFAS, was also associated with carpet status. We did not observe associations between carpet status or furniture with the phased-out PFOS. Although PFBS is thought to be used as a replacement for PFOS in Scotchgard (California Environmental Protection Agency, 2019; Glynn et al., 2012), we measured it too infrequently to include in our analyses. This may be because we had a relatively high detection limit in this study compared to other studies with PFBS (Young et al., 2020). Further, installation of carpet in these spaces may have occurred before the replacement of PFBS for PFOS in carpet treatments. We do not have information on the year carpet was installed. Since carpet has been shown to be a reservoir for semivolatile organic compounds (SVOCs) (Haines et al., 2020), it is surprising that we did not see consistent associations between carpeting and dust concentrations for other non-PFAS chemicals. However, the lack of association may be because we did not have information for when the carpets were introduced into each space; for example, it is likely that carpeting has been replaced, perhaps multiple times, since DDT was banned from use, meaning that DDT may be correlated to other components in the spaces or widely redistributed throughout the indoor space. Unlike the OCPs and PCBs in our study, it is likely that PBDEs and PFAS have current sources in furniture and carpeting in the spaces.

Consistent with previous studies of PBDEs and furniture, we found positive significant associations between BDE 100 and BDE 153 concentrations and years since last furnished and BDE 209 concentrations and number of pieces of upholstered furniture. Furniture, particularly older furniture, appeared to be a significant source of PBDEs in our

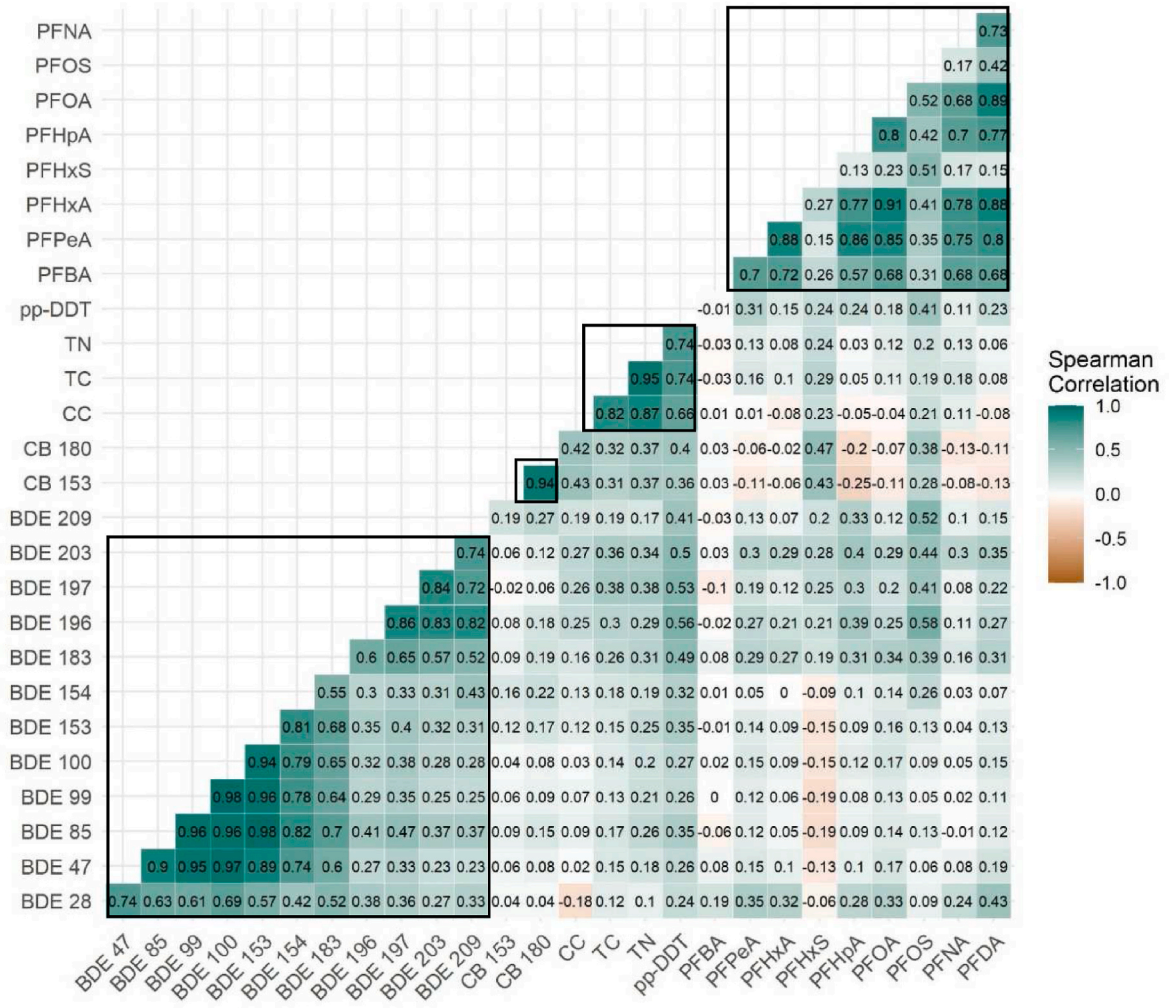

Fig. 2. Spearman correlation estimates for persistent chemicals measured in dust. Chemical groups indicated by black squares.

study's spaces. This finding is consistent with other studies that have found relationships between upholstered furniture and PBDE levels in college settings (Rodgers et al., 2020; Young et al., 2020; Dodson et al., 2017).

This is one of the first studies to investigate a mixture of PFAS, PBDEs, PCBs and OCPs in dust. Persistent chemicals may have similar structures and toxicological endpoints (*e.g.*, endocrine disruption, thyroid toxicity), potentially leading to additive or interactive health effects (Curtis et al., 2019; Dingemans et al., 2016; Lopez-Espinosa et al., 2012; Turyk et al., 2007). Therefore, the presence of persistent chemical mixtures in dust indicates that human health risk assessments focused on individual chemicals may be inadequate and should consider exposure to mixtures to be protective of health.

4.1. Limitations

This study evaluated the presence of PFAS and a mixture of four classes of persistent chemicals in non-residential institutional spaces and their association with a number of room characteristics. However, our study has several limitations. For example, there may be other important indoor sources of PFAS that we did capture, including use of carpet treatments, cleaning solutions, or other maintenance practices that may contain PFAS. Concentrations of PFOS were among the highest PFAS observed in our study, and given PFOS was not associated with carpeting or furniture, other sources of this chemical likely exist that we did not quantify. We also measured only a limited number of PFAS, and did not measure precursor chemicals that have been detected in indoor spaces, and only sampled dust from four colleges in New England; therefore, our finding may not be generalizable to colleges in different states and geographical regions. Further, our small sample size limits the statistical power and precision of our findings. Lastly, the room characteristics, particularly carpet status, may have been measured with error (e.g., carpets may have covered the majority of the space but not be considered wall-to-wall), though we would expect any misclassification to be non-differential and create a bias towards the null.

4.2. Implications

The presence of these four classes of persistent chemicals, years after their bans in many cases, illustrates the need to characterize chemical persistence before manufacture and widespread use occurs. For example, PFOS and PFOA were largely phased out by 2002 and 2010, respectively, and were found in 77–100% of samples, and DDT was banned in 1972, but was found in 93% of samples. There is scientific evidence for regulating chemicals based on persistence alone, since persistence is a chemical characteristic that has the potential to lead to cumulative exposures and health effects for decades or longer (Cousins et al., 2019a). Further, the case of PFAS has demonstrated the cost and technical hurdles associated with addressing contamination of persistent chemicals, emphasizing the importance of regulatory policies that prevent this kind of contamination before widespread exposure occurs. Applying a "persistence-sufficient" approach to PFAS and other

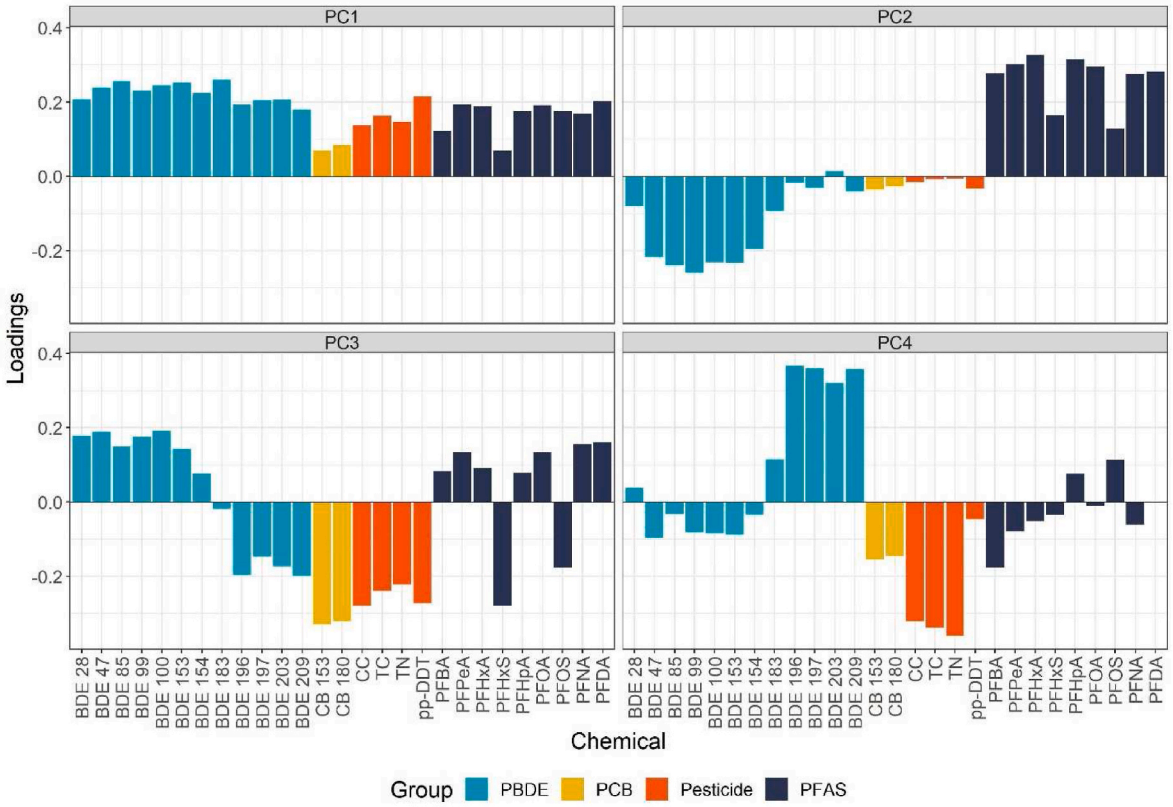

Fig. 3. Loadings of the first four principal components (PCs) from the PCA.

Table 2
Mutually adjusted associations between room characteristics and principal component scores for the first four principal components. Beta and 95% confidence intervals shown.[a,b]

	carpet	no. of upholstered furniture	years since last furnished
PC-1	2 (−0.4, 4.4)	−0.00042 (−0.0076, 0.0067)	0.051 (−0.078, 0.18)
PC-2	1.9 (0.19, 3.6)	−0.00077 (−0.0059, 0.0043)	−0.074 (−0.17, 0.018)
PC-3	−0.13 (−1.7, 1.4)	−0.0046 (−0.0092, 0.000032)	0.024 (−0.059, 0.11)
PC-4	0.57 (−0.6, 1.7)	0.0033 (−0.0002, 0.0068)	−0.026 (−0.089, 0.036)

[a] All chemical concentrations were log-transformed and z-standardized.
[b] The PCA was constrained to four principal components, which explained ~75% of the variance in the data.

manufactured chemicals would put an end to the repetitive cycle of allowing persistent chemicals to be produced without adequately considering multi-generational consequences (Cousins et al., 2019a; Balan et al., 2021).

Given the persistence and toxicity of PFAS that have already entered the environment, and will remain for hundreds of years, further exposures need to be prevented. For example, institutions should prioritize healthy materials during remodeling or building projects. There are many certification programs that contain criteria for using non-hazardous products (Goodwin Robbins et al., 2020). Another strategy is to eliminate non-essential (or avoidable) uses of PFAS (Cousins et al., 2019b; Kwiatkowski et al., 2020), including carpeting, paints, and cleaning mixtures, as has been outlined in the European Union's Chemical Strategy (Council of the European U, 2019). Our findings also underline the importance of regulating persistent chemicals at the class-level, as we found detectable concentrations for PFAS (PFOA, PFOS, and PFHxS) and other persistent chemicals (*e.g.*, DDT and PCBs) that have been banned or phased-out for decades. In addition, our findings support the need for risk assessment strategies that incorporate cumulative and additive effects from multiple chemical exposures. There is growing support for regulating PFAS as a class based on their common persistent properties (Cousins et al., 2019a, 2020; Balan et al., 2021; Kwiatkowski et al., 2020). Changes in chemical policy need to happen simultaneously at building, institutional, municipal, state, and national levels, and our findings support the need for safer chemicals in each of these arenas.

Funding sources

This work was supported by the John Merck Fund, the Fine Fund, the Hoffman Program on Chemicals and Health at Harvard T.H. Chan School of Public Health, and charitable donations to Silent Spring Institute.

Declaration of competing interest

The authors declare no competing financial interests.

Acknowledgments

This work was supported by the John Merck Fund, the Fine Fund, the Hoffman Program on Chemicals and Health at the Harvard T.H. Chan School of Public Health, and charitable donations to Silent Spring Institute. We thank the participating colleges and M. Dashko, L. Swetschinski, L. Borth, and K. Vilardi for assistance with sampling and C. Christia for help with the chemical analysis.

The research presented was not performed or funded by EPA and was not subject to EPA's quality system requirements. The views expressed are those of the author(s) and do not necessarily represent the views or the policies of the U.S. Environmental Protection Agency.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2021.112530.

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