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Publication Date

2016

Peer reviewed

Association between Serum Polybrominated Diphenyl Ether Levels and Residential Proximity to Solid-Waste Facilities

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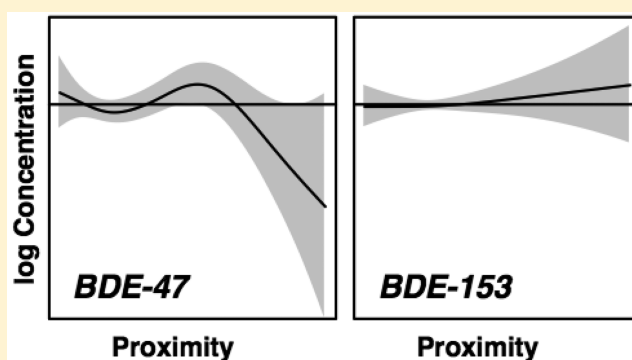
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S Supporting Information

ABSTRACT: As consumer products treated with polybrominated diphenyl ethers (PBDEs) reach the end of their life cycle, they often are discarded into solid-waste facilities, offering a potential reservoir for exposure. The likelihood of exposures to PBDEs by residents living near those sites rarely has been explored. This study collected blood samples from 923 female participants in the California Teachers Study in 2011–2013 and examined the association between participants' residential proximity to solid-waste facilities with potential release of PBDEs and serum levels of three congeners (BDE-47, BDE-100, and BDE-153). General linear regression analysis was used to examine the association, adjusting for age, race, body-mass index, neighborhood socioeconomic status, and urban residency. Compared to participants living >10 km from any selected site, those living within 2 km had 45% higher BDE-47 (95% CI: 5–100%) and BDE-100 (95% CI: 0–109%) levels, and those living between 2 and 10 km had 35% higher BDE-47 (95% CI: 0–82%) and 29% higher BDE-100 (95% CI: –9 to 82%) levels. No associations were found for BDE-153. Living close to some solid waste sites may be related to higher serum BDE-47 and BDE-100 levels. Studies with comprehensive exposure assessments are needed to confirm these initial observations.



are moving from widespread use to disposal into the solid-waste stream.

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) have been widely used as flame retardants in various products, including plastics, wire insulation, building materials, household and business furnishings, and automobiles.^{1,2} PBDEs can be emitted into the environment in each phase of their life cycle, from PBDE production to the use and final disposal of PBDE-treated consumer products. As illustrated in Figure S1,³ PBDEs are transported via multiple media, including air, soil, and water, and accumulate in the food chain. PBDEs have been detected in house dust, in both indoor and outdoor air, and in different food groups in many countries worldwide.⁴ PBDEs are also found in various waste streams, such as electronic waste, autoshredder waste and sewage sludge,⁵ and in landfill leachate.⁶ Because PBDEs are persistent and ubiquitous in multiple environmental media, human exposure can occur via multiple routes, including the ingestion of contaminated food, dust, and water; inhalation of gas, dust, and particles from indoor or outdoor air; and dermal absorption of dust or via contacts with various consumer products. The relative contribution of each of these routes has not been well-characterized and is likely changing as PBDE-laden products

are moving from widespread use to disposal into the solid-waste stream.

There is evidence that PBDE levels among Americans are much higher than those among Asians and Europeans,⁷ and Californians have higher PBDE levels than the rest of the U.S. population, likely due to California's furniture flammability standard.⁸ Accumulating evidence shows that human-body levels of PBDEs are associated with measured PBDE levels in house dust as well as the consumption of dairy products, meat, and fish.^{9–11} It has been estimated that dust and soil ingestion and dermal contact might explain 90% of a U.S. adult's daily exposure to total PBDEs.⁷

Laboratory studies have shown that PBDEs may have endocrine-disrupting properties and are associated with multiple neurobehavioral, developmental, and reproductive effects.^{12,17,18} Epidemiological studies, while limited, have also

Received: September 25, 2015

Revised: February 16, 2016

Accepted: February 24, 2016

Published: February 24, 2016

observed some human health risks associated with PBDE exposure.^{19,21,22}

Due to concerns about PBDEs' impact on human health and the environment, California banned the two most commonly used formulations, penta- and octa-BDEs in 2006,²³ and all uses of deca-BDE were required to end by the end of 2013.⁷ As a result, indoor exposures from the dust and off-gassing of PBDEs associated with the use of PBDE-laden consumer products are likely to become a less-important route of exposure, while outdoor exposures may become more predominant. Thus, evaluating whether people's body burden of PBDEs may be associated with residential exposure to PBDEs from solid waste disposal sites is of public health significance because it may provide important implications for solid-waste management to reduce future exposures to these chemicals.

A pair of studies have reported an association between body levels of PBDEs and potential residential exposures related to waste-disposal sites. One was conducted in China, comparing workers in an electronic-waste-dismantling region to nearby residents and the general population.²⁴ The other was conducted in Nicaragua among young people working at or living near a large municipal waste disposal site (or both).²⁵ Both of these studies, however, were designed to examine occupational exposures to PBDEs and compare them to exposures in nearby residents who may have also been occupationally exposed. To our knowledge, no studies to date have been designed to examine people's residential exposure from waste-disposal facilities among the general U.S. population without substantial occupational exposures. The objective of our study was to evaluate the association between residential proximity to solid-waste facilities and serum levels of PBDEs among a sample of California adult women.

MATERIALS AND METHODS

Study Population. The study population consisted of 923 participants drawn from the California Teachers Study (CTS), a prospective cohort study consisting of 133 479 female professional public school employees initiated in 1995–96 primarily to study breast cancer. A full description of the cohort is described elsewhere.²⁶ Participants for the current analysis consisted of women serving as controls in an ongoing breast cancer case control study nested within the CTS and a convenience sample of additional breast-cancer-free CTS participants, which targeted nonwhites to ensure racial and ethnic diversity. Participants were required to have no prior history of breast cancer, live in California, and complete a supplementary interviewer administered questionnaire at the time of the blood draw in 2011–2013. The use of human subjects was reviewed and approved by the California Health and Human Services Agency, Committee for the Protection of Human Subjects, and all participating centers' Institutional Review Boards.

Serum Collection. Blood was collected from participants during 2011–2013 by licensed phlebotomists, usually in the participants' homes. Specimens were kept on cool packs and, within several hours of collection, were spun down using portable centrifuges to separate the serum portion. Processed samples were then frozen and transported to the laboratory, where they remained frozen and stored at -20°C until thawed for the PBDE assays.

PBDE Analysis. Serum PBDEs were analyzed by the Environmental Chemistry Laboratory at the California Depart-

ment of Toxic Substances Control (Berkeley, CA). Samples were thawed and aliquoted for PBDE and lipids measurements. Automated solid-phase extraction (SPE; Biotage, Uppsala, Sweden) and gas chromatography–high-resolution mass spectrometry (GC–HRMS, DFS, ThermoFisher, Bremen, Germany) were used for the analysis of PBDEs.²⁷ Briefly, thawed serum samples (2 mL) were fortified with a panel of $^{13}\text{C}_{12}$ -labeled surrogate mix standards (Wellington Laboratories, Inc., Guelph, Ontario, Canada) and mixed well. Equal volumes (4 mL) of formic acid and water were added into each sample before loading on the SPE modules. Oasis HLB cartridges (3 cc, 500 mg, Waters Co., Milford, MA) and acidified silica (500 $^{\circ}\text{C}$ prebaked, manually packed, 3 cc) were used for the sample extraction and cleanup, respectively. The collected final eluates in hexane/dichloromethane (1:1) were concentrated in a TurboVap (Biotage, Charlotte, NC) and spiked with recovery standard. Standard reference material (SRM 1958, National Institute of Standards and Technology, Gaithersburg, MD) and bovine serum prespiked with known amounts of target analytes were used as QA/QC samples.

A small volume of sera from each sample was sent to Boston Children's Hospital for the measurement of total cholesterol and triglycerides by enzymatic methods.^{28,29} Cholesterol and triglycerides were used to calculate the lipid content based on Phillips' formula.³⁰ PBDE levels were then lipid-adjusted to produce values with units of ng/g lipid.

Although a total of 19 congeners were analyzed in the laboratory, only those with detection frequencies of 75% or more were included in the current analysis to minimize potential biases associated with imputing a high frequency of nondetectable levels. Detection frequencies ranged from 0.3% for BDE-17 to 87.4% for BDE-47. Although they did not meet the detection frequency criteria sufficient for inclusion in the current analysis, BDE-99 and BDE-28 were detected in a substantial proportion of study participants, with 42.3% and 43.0% detected, respectively. The three congeners for which $\geq 75\%$ were detected and therefore included in this study were: 2,2',4,4'-tetrabromodiphenyl ether (BDE-47), 2,2',4,4',6-pentabromodiphenyl ether (BDE-100), and 2,2',4,4',5,5'-hexabromodiphenyl ether (BDE-153). To be consistent with standard national biomonitoring data, we used a simple but common practice to replace concentrations below the laboratory limit of detections (LODs) with $\text{LOD}/\sqrt{2}$ before lipid adjustment. For the 923 participants, the means (ranges) of lipid-adjusted LODs for BDE-47, BDE-100, and BDE-153 were 4.90 (1.32–17.64), 1.11 (0.23–4.41), and 2.31 (0.50–8.82) ng/g lipid, respectively.

Identification of Solid-Waste-Disposal Facilities with Potential Emission of PBDEs. Information on solid-waste-disposal facilities throughout California was obtained from the database available on the web site of the Solid Waste Information System (SWIS) of California's Department of Resources Recycling and Recovery.³¹ The database is updated frequently. Data used in this study were downloaded on May 28, 2013. Although these data do not include any specific information about potential PBDE exposures, we selected for inclusion in our study only those facilities that were likely to serve as potential sources of PBDE exposure on the basis of the limited knowledge contained in the data set regarding the activities and materials handled at each SWIS facility. Because the blood samples in our study were collected between 2011 and 2013, serum PBDE levels were unlikely to be related to exposures that might have occurred 30 years ago, so we

Table 1. Lipid-Adjusted Serum PBDE Levels (ng/g Lipid) among 923 California Female Teachers

	N(%)	BDE-47 ^a			BDE-100 ^a			BDE-153 ^a		
		GM	95% LCI	95% UCI	GM	95% LCI	95% UCI	GM	95% LCI	95% UCI
all	923 (100%)	14.21	13.37	15.09	2.46	2.30	2.62	5.51	5.15	5.89
age at blood draw		<i>p</i> = 0.87			<i>p</i> = 0.48			<i>p</i> = 0.22		
40–49	68 (7%)	14.19	11.60	17.35	2.57	2.04	3.24	7.08	5.58	8.98
50–59	149 (16%)	14.14	12.28	16.29	2.36	2.00	2.78	5.90	4.93	7.06
60–69	359 (39%)	14.08	12.77	15.53	2.51	2.26	2.79	5.43	4.87	6.04
70–79	248 (27%)	14.54	12.96	16.31	2.48	2.21	2.79	5.20	4.61	5.86
80+	99 (11%)	13.95	11.20	17.37	2.26	1.78	2.87	5.12	4.02	6.51
race		<i>p</i> = 0.0075			<i>p</i> = 0.034			<i>p</i> = 0.28		
Asian/PI	92 (10%)	17.23	14.24	20.86	2.79	2.29	3.41	5.92	4.84	7.23
Hispanic	95 (10%)	15.42	12.85	18.50	2.61	2.18	3.13	5.84	4.84	7.05
non-Hispanic White	637 (69%)	13.43	12.50	14.43	2.32	2.15	2.51	5.29	4.87	5.74
Black	87 (9%)	16.93	13.59	21.10	3.13	2.50	3.92	6.37	5.05	8.04
other	12 (1%)	9.25	4.98	17.19	1.88	1.01	3.47	5.99	2.16	16.62
BMI (at baseline, kg/m²)		<i>p</i> < 0.0001			<i>p</i> < 0.0001			<i>p</i> = 0.99		
unknown	33 (4%)	11.60	8.19	16.44	2.10	1.45	3.03	5.17	3.66	7.31
17–24	544 (59%)	12.49	11.58	13.49	2.12	1.96	2.30	5.47	5.01	5.97
25–29	226 (24%)	16.83	14.91	19.01	2.98	2.61	3.41	5.70	4.94	6.57
30–54	120 (13%)	19.54	16.48	23.15	3.44	2.88	4.11	5.43	4.54	6.51
neighborhood SES (quintiles)		<i>p</i> = 0.0016			<i>p</i> = 0.006			<i>p</i> = 0.16		
unknown	81 (9%)	11.57	9.34	14.32	2.04	1.62	2.57	4.53	3.62	5.68
1st (lowest)	36 (4%)	14.94	10.49	21.28	2.77	1.94	3.94	6.33	4.40	9.11
second	76 (8%)	20.86	16.59	26.23	3.47	2.74	4.41	6.40	5.15	7.97
third	150 (16%)	13.95	11.90	16.36	2.57	2.17	3.03	5.55	4.70	6.55
fourth	252 (27%)	14.01	12.52	15.68	2.42	2.13	2.74	5.40	4.72	6.18
fifth (highest)	328 (36%)	13.86	12.60	15.24	2.32	2.09	2.57	5.56	4.97	6.24
neighborhood urbanization		<i>p</i> = 0.32			<i>p</i> = 0.39			<i>p</i> = 0.50		
rural	64 (7%)	11.94	9.46	15.06	2.00	1.55	2.57	5.33	4.10	6.92
urban cluster	30 (3%)	14.57	10.75	19.76	2.31	1.77	3.01	6.08	4.68	7.91
urban	829 (90%)	14.39	13.49	15.34	2.50	2.33	2.68	5.50	5.12	5.92
residential proximity to the closest SWIS facilities^b		<i>p</i> = 0.025			<i>p</i> = 0.025			<i>p</i> = 0.57		
≤2 km	196 (21%)	15.81	13.72	18.22	2.82	2.42	3.29	5.92	5.11	6.86
>2 to 10 km	690 (75%)	14.05	13.13	15.04	2.41	2.24	2.59	5.41	5.00	5.85
>10 km	37 (4%)	9.89	7.41	13.20	1.72	1.24	2.38	5.25	3.69	7.48

^aThe detection frequency for BDE-47, BDE-100, and BDE-153 was 87%, 77%, and 79%, respectively. *p* values were based on the Kruskal–Wallis test. ^bTrend *p* value for BDE-47, BDE-100, and BDE-153 was 0.013, 0.005, and 0.08, respectively. GM: geometric mean; LCI: lower confidence interval; UCI: upper confidence interval; PI: Pacific Islander; BMI: body mass index; SES: socioeconomic status.

excluded those closed before Jan 1, 1980. For the same reason, facilities designated as “preregulation” (i.e., sites that ceased operations prior to August 15, 1977, when solid-waste facility permits began to be required) were also excluded. There is no information on the close date for those facilities, so they were excluded not on the basis of specific close date but of their “preregulation” status. Because asbestos can be used as a kind of fire retardant, we think that asbestos-containing materials are less likely to have PBDEs added; thus, asbestos-containing waste-disposal facilities were excluded.

There was a total of 3186 facility entries listed in the SWIS database, and facilities that are not likely to emit PBDEs were excluded sequentially on the basis of the rationale presented above, as follows: (1) closed before Jan 1, 1980 (*n* = 572); (2) in planning stages when the database was updated or “clean-closed” with documented removal of solid waste (*n* = 74); (3) “pre-regulations” (ceased operations prior to August 1977, *n* = 296); (4) composting sites accepting nonhazardous vegetable, yard, and wood wastes (*n* = 397); (5) waste-tire sites storing, stockpiling, discarding, collecting, or processing waste tires (*n* = 8); (6) involved in the following activities (*n* = 93): asbestos-

containing waste disposal, land application, construction and demolition wood debris chipping and grinding, nonhazardous-petroleum-contaminated soil disposal, sealed-container transfer operation, wood-waste disposal or sludge drying, or served as septage ponds, drilling mud impoundments, or leachate evaporation ponds; and (7) duplications with the same facility address (*n* = 176). A total of 1570 facilities were included in our study, 864 (55%) of which were solid-waste landfills (disposal sites) and the remainder being transfer and processing facilities that were utilized to receive solid wastes; temporarily store, separate, convert, or otherwise process the materials in the solid wastes; or to transfer the solid wastes directly from smaller to larger vehicles for transport; and those facilities utilized for transformation.

Calculating Participants’ Residential Distance to Selected SWIS Facilities. Latitude and longitude coordinates of SWIS facilities were downloaded from the SWIS web site. Participants’ residential addresses at time of blood draw were standardized using address-cleaning software (ZP4, Semaphore Corp.) and geocoded against parcel and then street centerline databases, and their distances to selected facilities were

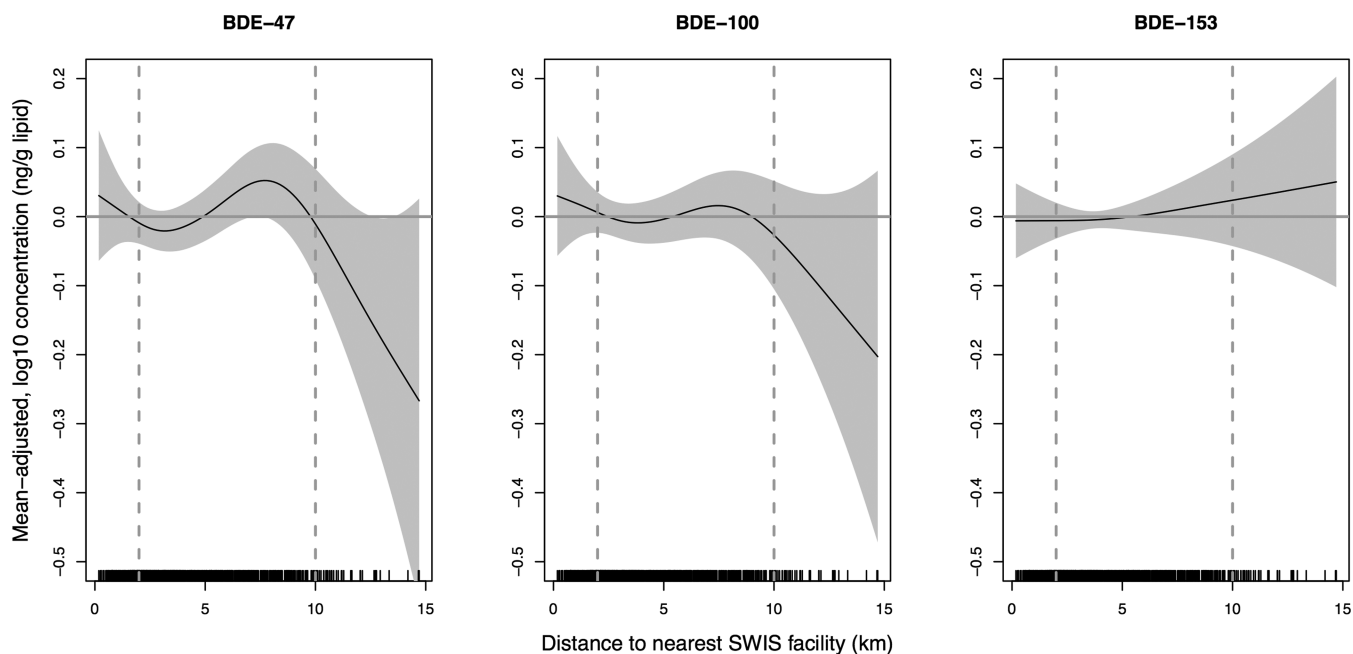


Figure 1. Serum PBDE congener levels vs residential proximity to nearest SWIS facility for 923 California female teachers. Note that for each congener, the panel shows (i) the result of an additive model regressing log concentration on smoothed distance to the nearest SWIS facility, smoothed age, and race; (ii) shading representing approximate 95% confidence intervals; (iii) a horizontal line at zero, representing the log-transformed overall geometric mean of the concentration; and (iv) vertical lines at 2 and 10 km. To reduce the effect of two outliers, we made the fit to proximities not exceeding 16 km.

calculated. All GIS was performed using ArcGIS 9.2 (ESRI, Redlands, CA).

Participants' Neighborhood Characteristics. Neighborhood socioeconomic status (SES) was derived by geocoding participants' residential addresses to U.S. Census 2000 block groups. Measures of block group SES were based on a previously developed method that incorporates census-based measures of income, occupation, and education at block group level.³² This variable was then categorized into quintiles, with the first quintile representing the lowest and the fifth quintile the highest summary measure of neighborhood SES across the 31 269 block groups in California in 2010. Each participant was assigned this composite measure of neighborhood SES on the basis of her geocoded block group.

Participants' residential addresses also were assigned an urbanization level according to the 2010 census urban classification scheme.³³ Participants were classified as living in an urban area (region of $\geq 50\,000$ people), an urban cluster (region of between 2500 and 50 000 people) or a rural area (region of <2500 people).

Statistical Analysis. Data analyses focused on assessing statistical relationships between lipid-adjusted serum PBDE concentrations and proximity to SWIS facilities while adjusting for important confounders. Analyses were conducted using SAS 9.3 (SAS Institute Inc., Cary, NC) and R.³⁴ The concentration distributions of the three congeners were highly skewed, with mean-to-median ratios around two (see Figure S2 for marginal density estimates and summary statistics for the congeners). Thus, all concentrations were log₁₀-transformed prior to statistical analysis. Although the distribution of proximity showed only a small amount of skew, there were two observations that appeared to be outliers. These two observations were eliminated prior to subsequent regression analyses when proximity was included as a continuous variable.

(See Figure S3 for a rug plot and density estimate for proximity.)

Our previous preliminary analysis of predictors of serum PBDE levels among 289 disease-free female California teachers showed that age, race, body mass index (BMI), and neighborhood SES were associated with participants' serum levels of some PBDEs.³⁵ Other studies have also shown that BMI³⁶ and race^{37,38} are associated with body PBDE levels. It has also been reported that PBDE levels in residential dust differ between homes located in urban, suburban, and rural areas.³⁹ Thus, we conducted descriptive data analyses and calculated the geometric means (GMs) and 95% confidence intervals (CIs) of the three congeners by levels of age, race, BMI, neighborhood SES, and urban residence.

To discover the general nature of the relationship between log concentration and proximity while adjusting for age and race, we performed exploratory analyses using the gam function in R's mgcv library. We fit additive models regressing log₁₀ concentration on the race and thin-plate smoothing spline (TPSS) functions of age and proximity, as well as augmented versions of those models that included other important regressors, such as BMI, neighborhood SES, and urbanization. Defaults were used for TPSS dimension and smoothness parameter selection. On the basis of the results of these exploratory analyses, as well as for a priori reasons, participants' residential proximity to the closest SWIS facility was included into the models as a three-level categorical variable. Specifically, for landfill emissions, 2 km has been reported as the likely limit of dispersion, with the possibilities of exposures over longer distances.⁴⁰ In addition, initial exploratory analyses indicated that the nature of the response changed at approximately 10 km. On the basis of these initial exploratory analyses, we classified participants' residential proximity to three levels: ≤ 2 , $>2-10$, and >10 km, with >10 km as the reference group.

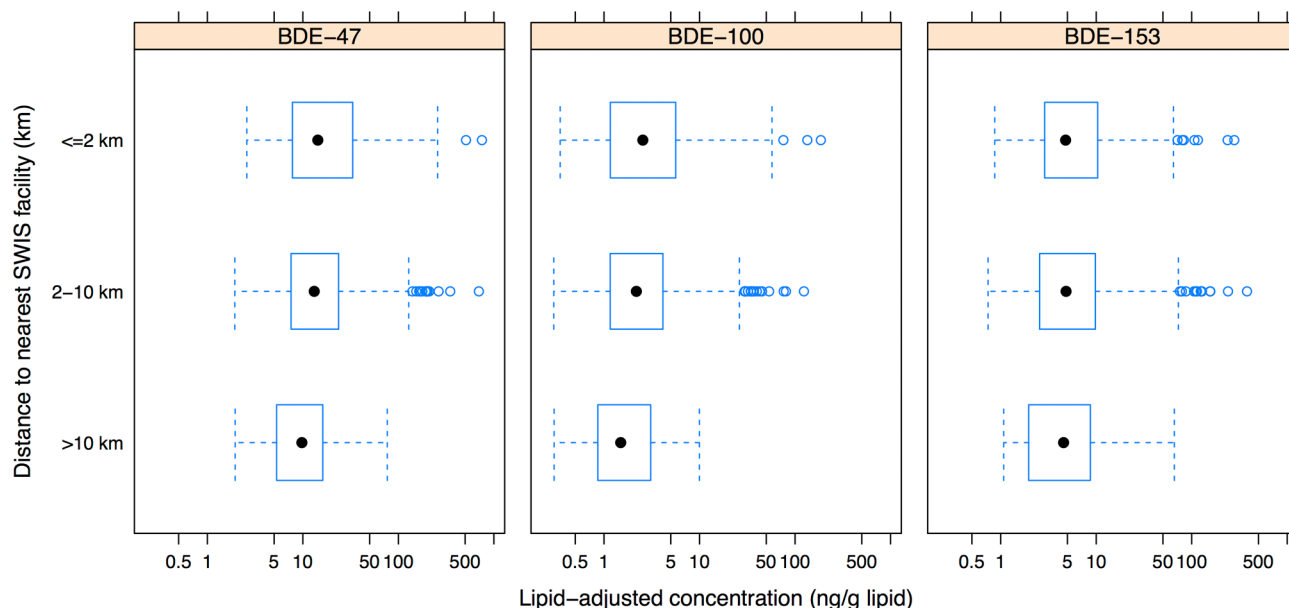


Figure 2. Distribution of serum PBDE concentrations by proximity to a SWIS facility. Each panel shows, on a log scale, an estimate of the marginal distribution of lipid-adjusted PBDE concentrations for one of the three congeners studied as a function of the distance in kilometers from the nearest SWIS facility.

Table 2. Association of Lipid-Adjusted Serum BDE-47, BDE-100, and BDE-153 Levels with Residential Proximity to SWIS Facilities: Estimates from General Linear Models Adjusting for Covariates among 923 California Female Teachers, 2011–2013^a

congener	proximity to SWIS facility	adjusted for age and race only			adjusted for age, race, BMI, neighborhood SES, and urbanization		
		estimate	95% CIs	p value	estimate	95% CIs	p value
BDE-47	≤2 km	1.58	1.15–2.19	0.008	1.45	1.05–2.00	0.032
	>2 to 10 km	1.41	1.05–1.91	0.024	1.35	1.00–1.82	0.060
	>10 km	1.00	–	–	1.00	–	–
BDE-100	≤2 km	1.62	1.15–2.29	0.009	1.45	1.00–2.09	0.050
	>2 to 10 km	1.38	1.00–1.91	0.055	1.29	0.91–1.82	0.149
	>10 km	1.00	–	–	1.00	–	–
BDE-153	≤2 km	1.12	0.79–1.66	0.510	1.15	0.79–1.66	0.491
	>2 to 10 km	1.02	0.72–1.45	0.909	1.02	0.72–1.45	0.866
	>10 km	1.00	–	–	1.00	–	–

^aLog₁₀-transformed concentrations of PBDEs were used in the regression analysis, and the estimates and 95% confidence intervals (\hat{y}) presented in this table were calculated via the equation: $y = 10^{\hat{x}}$, where x is the estimates from the regression analysis with log₁₀-transformed concentrations. CIs: confidence intervals; BMI: body mass index.

Analyses including only nonmovers (i.e., those who had the same residential addresses at baseline in 1995–1996 and at the time of blood draw) were also conducted to reduce potential misclassification due to address changes prior to blood draw.

RESULTS

Table 1 presents the lipid-adjusted serum PBDE levels for each of the three congeners by participant characteristics. Of the 923 participants included in this study, 82% were 50 to 79 years old at the time of blood draw, 69% were non-Hispanic white, 37% had BMI values of 25 kg/m² or greater, 63% lived in neighborhoods with relatively higher SES (the fourth and fifth quintiles), and 90% lived in urban areas. The detection frequencies for BDE-47, BDE-100, and BDE-153 were 87%, 77%, and 79%, respectively, and the geometric means (GMs) were 14.21, 2.46, and 5.51 ng/g lipid for these three congeners, respectively. Serum levels (log₁₀ transformed) of these three congeners were statistically significantly ($p < 0.0001$) correlated with each other, with a Pearson correlation coefficient of 0.92

between BDE-47 and BDE-100, 0.64 between BDE-100 and BDE-153, and 0.50 between BDE-47 and BDE-153.

For both BDE-47 and BDE-100, lipid-adjusted serum levels did not differ significantly by categories of age at blood draw or neighborhood urbanization, but they were higher among nonwhite, overweight, or obese participants and among those living in lower SES neighborhoods (second quintile of SES). In contrast, for BDE-153, no statistically significant differences were observed for any of the covariates evaluated.

Figure 1 shows an estimate of the relationship between levels of each of the three congeners and proximity to the closest SWIS facility while adjusting for age and race. It appears from the graphs that, for BDE-47 and BDE-100, the relationship is nonlinear and probably monotonic. In addition, the nature of the response seems to be divided into three regions. For distances less than approximately 2 km or greater than approximately 10 km, the levels appear to decrease with increased distance. However, the levels appear to be approximately constant for distances between 2 and 10 km,

although the variability in levels for a given distance is sufficiently great that no definitive conclusion about dose–response can be ascertained. When the distances are categorized into three levels using the above thresholds (see Figure 2), levels of BDE-47 and BDE-100 (but not BDE-153) differed by participants' residential proximity to closest SWIS facilities, with higher levels observed at closer distances (trend $p = 0.013$ and 0.005 , respectively).

Table 2 shows the results from multivariable linear regression models estimating the association between lipid-adjusted serum PBDE levels and residential proximity to SWIS sites. When adjusting for age, race, BMI at baseline, and neighborhood SES and urbanization, compared to participants living more than 10 km away, those living within 2 km or between 2 and 10 km of any SWIS facility had 45% higher (95% CI: 5–100%) and 35% higher (95% CI: 0–82%) lipid-adjusted serum BDE-47 levels, respectively, and 45% higher (95% CI: 0–109%) and 29% higher (95% CI: –9–182%) BDE-100 levels, respectively. No difference was observed for BDE-153 levels related to participants' residential proximity to SWIS facilities. Adjusting for age and race alone also showed similar results, although the magnitudes of differences slightly increased and the p values slightly decreased. For BDE-47 and BDE-100, the adjusted R^2 was approximately 1% for age- and race-adjusted fits and 5% for completely adjusted fits. For BDE-153, the adjusted R^2 was essentially zero.

Regression analysis restricted to nonmovers ($n = 531$) showed similar results to those for all participants but with higher p values, a likely reflection of the smaller sample size (see Table S1). Analyses stratified by type of SWIS facility (e.g., disposal sites or transfer and transport sites) yielded similar results to our analyses considering all types of SWIS facilities combined with no evidence of differences across the two types of SWIS facilities (data not shown). No significant linear relationship between log-transformed lipid-adjusted serum levels and residential proximity to SWIS facilities was observed when proximity was included as a continuous variable (data not shown).

DISCUSSION

To our knowledge, our study is the first U.S. study specifically designed to examine the potential impact of solid-waste-disposal landfills on nearby residents' body-burden PBDE level. Our study showed that participants living within 10 km, especially those living within 2 km of any SWIS facility, had higher serum levels of BDE-47 and BDE-100 (but not BDE-153) than those living >10 km away. Our results, although not without limitations, suggest that solid-waste-disposal sites may be an important source of PBDE exposures to the general population living in close proximity to such sites.

A pair of studies have reported limited evidence on the potential impact of waste disposal sites on nearby residents' body-burden levels of PBDEs; both were conducted outside of the United States, and both focused on workers' exposure compared to nearby residents. Qu et al. reported serum PBDE levels among workers of an electronic-waste-dismantling region in China, comparing levels to residents living about 50 km from the region, and among the general population. They found that workers had higher serum levels of some PBDE congeners, such as BDE-47, BDE-100, and BDE-209 than those of residents living about 50 km from the electronic waste dismantling region, who, in turn, had higher levels than the general population.²⁴ Athanasiadou et al. assessed PBDE levels

among children (age 11–15 years old) working or living at or nearby a large municipal-waste-disposal site in Managua, Nicaragua, where electronic waste was rarely found. Adjusting for fish consumption, they found that children working and living at the disposal area had serum levels 20–50 times higher for BDE-28, BDE-47, BDE-66, and BDE-99 than those who neither lived nor worked in the area. Children who were living in a nearby area (proximity to the waste disposal site was not specified) but had never worked at the disposal area had PBDE levels that did not differ from those of children living in a remote urban area about 10–20 km away from the disposal area.²⁵

Our results suggest that serum levels of both BDE-47 and BDE-100 may be associated with close residential proximity to solid-waste sites. Municipal solid waste landfills are significant environmental reservoirs for PBDEs, with an estimated 1700 tons of PBDEs disposed of in U.S. landfills in 2005 alone.⁷ PBDEs in outdoor air can be present in both the vapor phase and particle phase, with higher brominated BDEs more likely to be in particle phase. Penta- and lower brominated BDEs are predominant in the vapor phase and can travel over 1000 km; deca-BDE is exclusively in particle phase and can travel over 500 km, while other PBDEs are distributed in both particle and vapor phases.⁷ Studies have detected PBDEs from leachates⁴¹ and soil⁴² in the vicinity of landfills, which can contaminate both surface water and groundwater. Because PBDEs can be released from the landfills and contaminate nearby air, soil, and water and accumulate in plants, animals and fish,³ nearby residents can be exposed to PBDEs via multiple routes (Figure S1). These exposure routes may include the inhalation of outdoor gases or particles emitted from the site; of contaminated indoor air via soil, leachates, and gas migration; ingestion of drinking water obtained from private wells contaminated by leachates; bathing and washing in contaminated water via skin contact or inhalation of evaporated volatile compounds; skin contact with contaminated soil or inhalation of evaporation from soil; and recreational use of areas close to landfill or contaminated land.⁴³ Studies have shown that PBDE levels in the human body are highly correlated with PBDE concentrations in house dust,^{7,9,11,44} which are mostly from indoor release of PBDEs from consumer products. As these products stop being manufactured, distributed, or used and more are disposed in landfills than are used in daily life, indoor emissions will likely pose proportionately less impact on an individual's body levels of PBDEs, while outdoor emission sources such as solid-waste landfills may play an increasingly important role among those living in close proximity to such sites as well as posing as potential reservoirs for contamination of the food chain.

It is challenging to comprehensively assess people's exposure from landfills. Several factors may impact nearby residents' exposures to PBDEs from landfills, such as how the landfills are managed, the specific products and processing activities involved, whether people grow vegetables or raise animals for personal consumption in their vicinity, whether they consume locally produced food, and whether they use contaminated private or public water supplies. Unfortunately, information on such factors was not available for this study. The low R^2 values of our regression models indicate that there are indeed other explanatory factors not captured by our analyses.

It is important to note that our results were not particularly robust to alternative exposure specifications. Regression analyses that modeled participants' residential proximity as a

continuous variable or as a categorical variable with 11 levels (<1 km, 1–2 km with the increment of one kilometer until 9–10 km, and >10 km) yielded no evidence of a linear gradient of increasing serum levels with decreasing proximity. The cut-point of 2 km used in our final analysis to define the “high exposure” category, however, was chosen a priori on the basis of the World Health Organization’s assessment of likely dispersion distances for landfill-related exposures.⁴⁰ Furthermore, initial exploratory analyses of our own data suggested that the cut-points of 2 and 10 km reflected change points in the dose–response relationships, providing empirical support for our a priori chosen cutpoints. The absence of a linear gradient may be due to considerable measurement error (proximities were calculated for a geographic point assigned to the facility’s address, which may or may not capture the exact location of the landfill and does not take into account the size or contents of the landfill). The lack of a linear gradient may also be a function of our inability to account for the likelihood that exposures to PBDEs from landfills may occur via multiple exposure routes and may be influenced by multiple modifying factors.

The fact that we see an association with proximity to solid waste sites for BDE-47 and BDE-100 but not BDE-153 may be a function of their different uses. There were three major commercial formulations of PBDEs: penta-, octa-, and deca-BDEs.⁷ The penta-BDE formulation consists of 25–47% of BDE-47, 35–50% BDE-99, 6–10% BDE-100, and 3–5% BDE-153 by mass, and it was mostly used in flexible polyurethane foam materials in carpet cushions and furniture. The octa-BDE formulation consists of 5–10% of BDE-153, 40% BDE-183, 21% BDE-197, and 5–35% of BDE-203 by mass, and it was used primarily in hard plastics for the manufacture of casings to television sets, computers, hairdryers, automotive parts, etc. These two formulations were phased out in California in 2006. Both BDE-47 and BDE-100 were constituents of the penta-BDE formulation, while BDE-153 was present in both formulations; this may explain the high correlation observed between participants’ serum levels of BDE-47 and BDE-100 and the only moderate correlations between these two and BDE-153. Sandanger et al. also observed a strong correlation between blood BDE-47 and BDE-100 but not with BDE-153 among Canadian women.³⁸ It should be noted that higher-brominated BDEs can degrade to lower-brominated BDEs in both the environment and in animal and human bodies;⁷ thus, it is challenging to determine which congeners people may have been originally exposed to.

Because household dust is probably currently the major source of exposure for the general population, the association between people’s serum level of PBDEs and exposure to outdoor sources such as SWIS facilities in our study might be expected to be modest. The large sample size of our study provided sufficient statistical power to detect potential associations. Another particular strength of our study stems from the fact that all participants were from a single occupation (i.e., teachers) and therefore are unlikely to have significantly different workplace exposure to PBDEs, reducing the potential confounding by occupational exposures. Our analyses, however, do have a number of limitations that merit discussion. Although the residential location of CTS cohort members is geographically representative of the statewide population, given the convenience sample upon which the current analyses are based, the applicability of our results to the general population is difficult to know. Another limitation of our study is that we

were unable to adjust for other exposure sources such as diet and household dust in our analyses. It is possible that participants living closer to SWIS facilities could have greater PBDE exposures from household, dietary, or other routes of environmental exposures, including other types of industry that may be located near solid-waste facilities. Furthermore, although we were able to adjust for neighborhood SES, there may be some residual confounding due to the lack of individual measures of SES. It also should be noted that the selection of solid-waste disposal sites from the SWIS website was based on our best knowledge of products with potential PBDE additives, and the amount of PBDE emissions from selected sites was not known. Also, although landfills can release PBDEs into the environment and contaminate air, water, and soil, the exposure routes related to landfills remain unknown. Given these uncertainties in exposure assessment, our study results should be interpreted with caution. Studies with comprehensive exposure assessments are needed to confirm the association between residential proximity to SWIS facilities and elevated serum BDE-47 and BDE-100 levels to clarify the potential impact of solid-waste disposal sites on people’s exposure to PBDEs. As PBDE-contaminated products are phased out of use and enter the waste stream over the course of the next few decades, such exposure sources could have important public health implications in the future that could be controlled by improved waste management and control.

■ ASSOCIATED CONTENT

📄 Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: [10.1021/acs.est.5b04715](https://doi.org/10.1021/acs.est.5b04715).

Table S1: association of lipid-adjusted serum BDE-47, BDE-100, and BDE-153 levels with residential proximity to SWIS facilities among residentially stable California female teachers (nonmovers, $n = 531$), 2011–2013: estimates from general linear models adjusting for covariates. Figure S1: human exposure to PBDEs related to the PBDE life cycle. Figure S2: distribution of serum PBDE concentrations (ng/g lipid) among 923 California female teachers. Figure S3: distribution of distance (km) to nearest SWIS facility among 923 California female teachers. (PDF)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This research was supported by funds provided by the Regents of the University of California, California Breast Cancer Research Program, grant no. 16ZB-8501, and the National Cancer Institute, grant R01 CA77398. The opinions, findings, and conclusions herein are those of the authors and do not necessarily represent those of the Regents of the University of California or any of its programs. We express our appreciation to all the participants in the California Teachers Study and to the researchers, analysts and staff who have contributed so much for the success of this research project. We also thank Minhthu Le for administrative support, Christine Duffy for

overseeing field data and biospecimen collection, and the California Teachers Study Steering Committee members who are responsible for the formation and maintenance of the cohort within which this study was conducted but who did not directly contribute to the current paper: Hoda Anton-Culver, Jessica Clague deHart, Christina A. Clarke, Dennis Deapen, Pamela Horn-Ross, James V. Lacey Jr, Yani Lu, Huiyan Ma, Susan L. Neuhausen, Hannah Park, Rich Pinder, Fredrick Schumacher, Sophia S. Wang, and Argyrios Ziogas.

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