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Concentrations and risks of *p*-dichlorobenzene in indoor and outdoor air

Abstract *p*-dichlorobenzene (PDCB) is a chlorinated volatile organic compound that can be encountered at high concentrations in buildings owing to its use as pest repellent and deodorant. This study characterizes PDCB concentrations in four communities in southeast Michigan. The median concentration outside 145 homes was 0.04 $\mu g/m^3$, and the median concentration inside 287 homes was 0.36 $\mu g/m^3$. The distribution of indoor concentrations was extremely skewed. For example, 30% of the homes exceeded 0.91 $\mu g/m^3$, which corresponds to a cancer risk level of 10^{-5} based on the California unit risk estimate, and 4% of homes exceeded 91 $\mu g/m^3$, equivalent to a 10^{-3} risk level. The single highest measurement was 4100 $\mu g/m^3$. Estimates of whole-house emission rates were largely consistent with chamber test results in the literature. Indoor concentrations that exceed a few $\mu g/m^3$ indicate the use of PDCB products. PDCB concentrations differed among households and the four cities, suggesting the importance of locational, cultural, and behavioral factors in the use patterns of this chemical. The high PDCB levels found suggest the need for policies and actions to lower exposures, for example, sales or use restrictions, improved labeling, and consumer education.

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Practical Implications
Distributions of *p*-dichlorobenzene concentrations in residences are highly right-skewed, and a subset of houses has very elevated concentrations that are equivalent to an excess cancer risk of 10^{-3} or higher based on the California unit risk effect estimate. House-to-house variation is large, reflecting differences in use practices. Stronger policies and educational efforts are needed to eliminate or modify indoor usage practices of this chemical.

Introduction

p-dichlorobenzene (PDCB) is a chlorinated volatile organic compound (VOC) that is widely used in essentially pure form (>99.8%) as a repellant against snakes, rats, mice, squirrels, bats, and insects, as a deodorizer for toilets, urinals, and diaper pails, as an insecticidal fumigant, and as an air freshener (ATSDR, 2006; Kelly, 2009; National Toxicology Information Program, 1993; Wilhide and Fletcher, 1995). When used as moth repellents, PDCB-containing products (in the form of crystals, flakes, or cakes) are typically placed in closed drawers, closets, and plastic bags where clothes, blankets, and other goods are stored. As a deodorizer, PDCB is often placed in a toilet, diaper pail, bathroom, attic, basement, garage, pet cage, vehicle, or other location where odor is a concern. There are no known natural sources of PDCB (IARC, 1999a). Outdoor emission sources include the volatilization of consumer and commercial products containing PDCB, waste sites, and manufacturing facilities (ATSDR, 2006). PDCB is relatively stable in the environment compared to other VOCs, and its estimated atmospheric half-life is 14–31 days (Howard, 1989; Mackay et al., 1992). Outdoor concentrations are usually low, generally below 1 $\mu g/m^3$ (Hultin et al., 2010; Johnson et al., 2010; Sexton et al., 2004; Simon et al., 2005; Weisel et al., 2005). In contrast, PDCB is commonly detected in indoor air, often at moderate to high concentrations. Exposures occur in both residential and occupational settings, including the manufacture of polyphenylene sulfide resins, deodorants,

mothballs, dyes, pharmaceuticals, and agricultural products (ATSDR, 2006). PDCB also has been found in finished drinking water, surface water, groundwater, soil, sediments, in meats owing to its use in deodorant blocks in animal stalls (ATSDR, 2006; Environment Canada, 1993), and in honey and royal jelly due to its use as an insecticide for empty beehives and bee houses stored indoors (ATSDR, 2006; Environment Canada, 1993; Tananaki et al., 2009).

The widespread use of products containing PDCB suggests the importance of understanding the emissions, concentrations, exposures, and health risks associated with this chemical, especially in indoor environments that have the highest potential for exposure. Exposure can be assessed by measuring airborne concentrations and PDCB metabolites in blood, urine, adipose tissue, and breast milk (Aronson et al., 2007; ATSDR, 2006). Exposure has been associated with several adverse effects. Inhalation exposure has produced malignant tumors in the livers of mice and hyperplasia in the kidneys of rats (Aiso et al., 2005). Parenteral exposure (subcutaneous and intraperitoneal) reduced sperm production and had anabolic–androgenic effects in rats and mice (Takahashi et al., 2011). In adult humans, elevated inhalation exposure has been linked to increased white blood cell counts (Hsiao et al., 2011) and decreased pulmonary function (Elliott et al., 2006). PDCB has been classified as possibly carcinogenic to humans (Group 2B) (IARC, 1999b). California lists PDCB as a possible human carcinogen and in 1994 assigned an inhalation cancer unit risk estimate (URE) of 1.1×10^{-5} per $\mu g/m^3$ (California OEHHA, 2009). The US Environmental Protection Agency considers PDCB to be a low-risk pesticide because of the lack of evidence for carcinogenicity, and it lists a non-cancer chronic inhalation reference concentration (RfC) of 800 $\mu g/m^3$ (USEPA, 2010). The US Occupational Safety and Health Administration (OSHA) permissible exposure limit is 450 mg/m^3 as an 8-hour time-weighted average concentration (OSHA, 2012). For general indoor air, Japan has specified a guideline value for lifetime exposure of 240 $\mu g/m^3$ (Japanese Ministry of Health Labour and Welfare, 2001).

The uses and exposure patterns of PDCB in buildings are very similar to those of naphthalene, which is employed for many of the same uses, and also sold in a solid form that sublimates rapidly (Batterman et al., 2012a; Jia and Batterman, 2010). Both chemicals have been identified as priority pollutants in residences (Logue et al., 2011). In addition to its use as a deodorizer and repellent, naphthalene is also a product of incomplete combustion and a component of gasoline and gasoline vapor. Consequently, naphthalene tends to be ubiquitous, for example, detected wherever combustion occurs or gasoline is stored. In contrast, PDCB levels can be very low or undetectable levels in buildings where this chemical is not used.

This paper provides current information on PDCB concentrations and risks in indoor and outdoor settings in four Michigan cities. We characterize indoor levels in nearly 300 homes, estimate sources of variability using variance proportions, examine high-end concentration distributions, discuss spatial trends of ambient concentrations, derive composite emission rates from houses, and estimate risks.

Methods

Sampling sites

p-dichlorobenzene concentrations were monitored in 287 households in four southeast Michigan, USA communities: Ann Arbor (AA), a largely suburban and affluent community of 113 934; Ypsilanti (YP), an economically diverse and urbanized town of 19 435 with more commercial activity; Dearborn (DB), population 98 153; and Detroit (DT), population 713 777 (The U.S. 2010 Census). The latter two cities are fairly densely populated industrialized cities that have relatively low household incomes and education levels. Households were recruited using several methods. Random sampling using telephone dialing and snowball recruitment methods garnered 161 households (65, 35, and 61 in AA, YP, and DB, respectively) as part of an exposure study (Jia et al., 2008a,b). In DT, as part of a community-based participatory research (CBPR) study on asthma, 126 households were recruited using questionnaires distributed to caregivers at various venues (e.g., schools and community fairs) aimed at obtaining households in which lived a child (6–12 years old) with symptoms or medication use consistent with persistent asthma (Parker et al., 2008). Informed consent and procedures approved by the University of Michigan Institutional Review Board were followed.

Information on housing, smoking, family characteristics, hobbies, and other factors potentially associated with exposure in each household was obtained using questionnaires administered to the head-of-household or child caregiver, and building walk-through surveys conducted by our technicians. The latter used a standardized form to note each home's characteristics and condition, for example, type of heating and cooling system, presence of attached garages, and potential emission sources, for example, incense and room deodorizers. These surveys did not inquire about uses and application rates of PDCB products, as the PDCB measurements were intended to indicate the use of these products.

Residences were monitored in at least two seasons. AA and YP residences were visited in summer 2004 and winter 2005; DB residences in fall 2004 and spring and summer 2005. In DT, households entered the study

between March 2009 and February 2010, and 92% of homes had two or more seasonal visits (11, 17, 87, and 12 homes had 1, 2, 3, and 4 visits, respectively). Monitoring in AA, YP, and DB included simultaneous indoor and outdoor sampling. Indoor samplers were deployed in the living room, and outdoor samplers at a location close to the house, for example, the backyard, at most (90%) homes. Each location used duplicate or triplicate samples. In DT, indoor samplers collected single or duplicate samples in the living room, and duplicate samples in the child's bedroom. Outdoor concentrations were not monitored in DT.

p-dichlorobenzene and other VOCs were measured using passive thermal desorption tube samplers over 3- to 7-day periods in each home (Batterman et al., 2002). Samplers were deployed at breathing height and away from windows, doors, obvious sources of potential contaminants, corners and other potentially stagnant areas, and out of the reach of children. Except for the variance analyses (described later), measurements at different locations in a residence were averaged, and seasonal measurements were averaged to obtain long-term concentration estimates. Thus, each observation is an average of replicates, two rooms for the DT homes, and two or more seasons. Overall, we collected a total of 1439 valid indoor samples and 478 valid outdoor samples. (Blanks and samples that failed because of sampling or analysis issues are omitted from these counts.)

Volatile organic compound analyses were performed by an automated thermal desorption-gas chromatography–mass spectrometry (ATD-GC-MS) system (Jia et al., 2006). Given the high PDCB levels occasionally encountered indoors, our 7-point calibration (equivalent to concentrations from 0.2 to 200 $\mu g/m^3$) was extended to 500 $\mu g/m^3$. Linearity remained excellent, for example, the R^2 was 0.996, and the relative standard deviation of duplicate measurements remained within 9%. Quality assurance (QA) measures included the use of standard operating protocols, weekly collection and analysis of blanks, regular flow checks, quarterly calibrations, and duplicate or triplicate samples. The method detection limit (MDL) was < 0.02 $\mu g/m^3$, and the replicate precision was $\leq 20\%$. Non-detects were set to 1/2 MDL.

Data analysis

To understand the sources of variation, random effects models were used to apportion the variance of the PDCB concentrations. Following Jia et al., 2011, five variance components were used: seasonal variability (e.g., variance between seasons); measurement uncertainty (variance between replicates); and three types of spatial variability: variances between cities; between residences; and within residences. The latter was estimated only for DT and specifically between bedrooms and living rooms. The variance analysis was performed for indoor (AA, YP, DB, and DT) and outdoor (AA, YP, and DB) concentrations. Because the random effects model assumes normality and PDCB concentrations followed log-normal distributions, analyses used log-transformed data.

We also used generalized extreme value (GEV) distributions to fit extreme PDCB and naphthalene concentrations, defined as values above the 90 percentile. Goodness-of-fit was evaluated using Anderson–Darling tests. Bivariate analyses examined PDCB and naphthalene levels in the residences, including contingency tables with quartile groupings for each pollutant, scatterplots, and correlations.

Emission rates from PDCB products can be estimated in several ways. As noted earlier, emission rates can be determined using chamber tests. While such tests can be well controlled, results may not reflect in-use or representative emission rates. Alternatively, emissions can be calculated for the house by considering the entire residence as a 'test chamber':

$$E = \lambda V(C_{in} - C_{out}) \quad (1)$$

where E = emission rate of the house ($\mu g/h$), λ = house air exchange rate (per h), V = house volume (m^3), and C_{in} and C_{out} = indoor and outdoor concentrations of PDCB ($\mu g/m^3$), respectively. These calculations used the median house volume V (360 m^3) and median air exchange rate λ (0.57/h) estimated for the DT homes (Batterman et al., 2012b; Du et al., 2011). Outside PDCB concentrations C_{out} in DT were not measured; thus, the median level in AA, YP, and DB was used. Air exchange rates in AA, YP, and DB were not measured and, as shown later, DT was the community with the most frequent use of PDCB. For the indoor concentration C_{in}, the 90th percentile concentration in DT was selected to portray residences using PDCB products.

Excess lifetime cancer risks were estimated using the URE from California, that is, 1.1×10^{-5} per $\mu g/m^3$ (California OEHHA, 2009).

The variance components analyses used PROC NESTED in SAS (v9.1.3; SAS Institute, Cary, NC, USA). The GEV analyses used GEV in R (v2.14.1, R Development Core Team, 2008).

Results

Indoor concentrations and comparison to the literature

p-dichlorobenzene was detected in nearly all (95%) homes. The residence-average PDCB concentrations ranged from detection limits to 2100 $\mu g/m^3$, and the single highest concentration was 4200 $\mu g/m^3$ (Table 1). The mean and median indoor PDCB concentrations were 21 and 0.36 $\mu g/m^3$, respectively. Medians varied among the four cities (Kruskal–Wallis test, $P < 0.01$)

Table 1 Statistics of indoor and outdoor concentrations of *p*-dichlorobenzene in four Michigan cities, both separately and combined ('all')

	Residence-based statistics					Visit-based statistics				
Location and statistic	AA	YP	DB	DT	All	AA	YP	DB	DT	All
Indoors										
Sample size	65	35	61	126	287	98	59	87	345	589
Detection frequency (%)	83	97	93	98	94	85	93	94	98	95
Concentration ($\mu g/m^3$)										
Mean	1.40	9.60	4.20	41.00	21.00	1.70	11.00	3.80	31.00	20.00
Standard deviation	6.40	35.00	20.00	210.00	140.00	10.00	44.00	18.00	250.00	190.00
Median	0.09	0.16	0.31	0.65	0.36	0.07	0.13	0.32	0.56	0.38
90th percentile	2.20	7.60	3.90	26.00	7.00	1.60	11.00	3.40	7.80	6.10
95th percentile	4.70	96.00	13.00	170.00	46.00	4.70	82.00	13.00	38.00	21.00
99th percentile	51	180	150	860	430	100	290	150	850	500
Maximum	51	180	150	2100	2100	100	290	150	4200	4200
Outdoors										
Sample size	53	35	57	–	145	85	59	81	–	225
Detection frequency (%)	87	100	91	–	92	79	92	90	–	86
Concentration ($\mu g/m^3$)										
Mean	0.03	0.10	0.13	–	0.09	0.04	0.11	0.14	–	0.09
Standard deviation	0.03	0.29	0.17	–	0.18	0.04	0.36	0.25	–	0.24
Median	0.03	0.03	0.08	–	0.04	0.02	0.03	0.06	–	0.03
90th percentile	0.07	0.09	0.34	–	0.17	0.09	0.11	0.30	–	0.16
95th percentile	0.10	0.56	0.39	–	0.34	0.14	0.83	0.49	–	0.30
99th percentile	0.14	1.70	0.98	–	0.98	0.18	2.50	1.90	–	0.89
Maximum	0.14	1.70	0.98	–	1.70	0.18	2.50	1.90	–	2.50

'Residence-based' statistics are the average of multiple visits at each residence. 'Visit-based' statistics are the average of multiple replicates and locations at each residence, but do not average across seasons. Indoor concentrations in Detroit (DT) use average of measurements in living rooms and bedrooms. Indoor concentrations in Ann Arbor (AA), Ypsilanti (YP), and Dearborn (DB) use average of measurements in living rooms. Outdoor concentrations use measurements near participant homes in AA, YP, and DB.

and were ranked as DT > DB > YP > AA. These measures of central tendency of PDCB concentrations fall within ranges reported in most of the other residential studies, for example, 1.2–69 $\mu g/m^3$ for means and 0.2–1.9 $\mu g/m^3$ for medians (Aronson et al., 2007; ATSDR, 2006; Jia et al., 2008c; Kinney et al., 2002; Logue et al., 2011; Sax et al., 2004; Sexton et al., 2004; Wallace et al., 1985, 1988; Weisel et al., 2005). An earlier residential study in DT in 2006 reported a similar mean concentration (17 $\mu g/m^3$, $N = 41$) (Johnson et al., 2010). Generally, comparable levels have been seen in studies using personal monitoring, including the nationally representative 1999–2000 US National Health and Nutrition Examination Survey (NHANES) study, which found median levels of 1.7 $\mu g/m^3$ ($N = 665$) among adults (Jia et al., 2008c), and an older national US study conducted around 1988, which reported a median personal level of 2.5 $\mu g/m^3$ ($N = 1650$) and a median indoor concentration of 1.4 $\mu g/m^3$ [$N = 2121$; (ATSDR, 2006)].

Distributions of indoor PDCB concentrations were strongly right-skewed with skewness coefficients from 4.4 to 8.2 depending on the city (Figure 1), and large differences between means and medians (Table 1). Indoor PDCB concentrations were sometimes extremely elevated. Five homes in DT (4%) exceeded the RfC (800 $\mu g/m^3$) in one seasonal visit, although levels in these homes were not high in other seasons tested. As noted, the single highest (7-day) measurement was

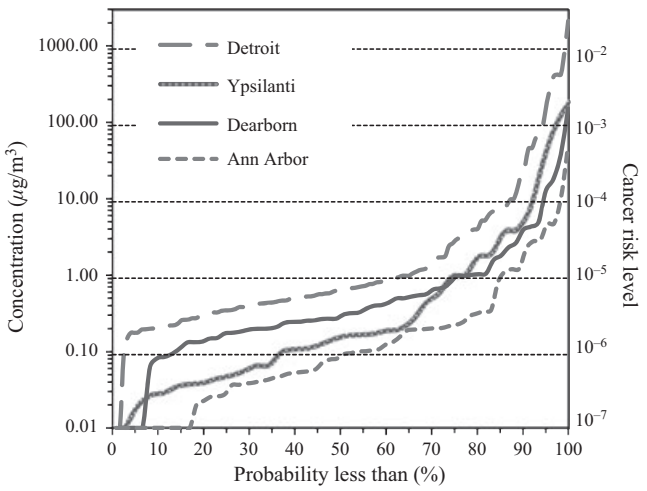

Fig. 1 Distributions of indoor *p*-dichlorobenzene concentrations (left axis), and individual excess cancer risk levels (right axis) in the four study cities ($N = 287$). Uses multiseason whole-house average. Cancer risks based on unit risk estimate of 1.1×10^{-5} per $\mu g/m^3$ (California OEHHA, 2009)

4200 $\mu g/m^3$. When averaged across rooms and seasons, this DT house also had the highest concentration (2100 $\mu g/m^3$). These levels slightly to greatly exceed those previously reported, for example, 4000 $\mu g/m^3$ (The Health Effects Institute, 2008), 1500 $\mu g/m^3$ (*m*- and PDCB combined) (Wallace et al., 1985), 1790 $\mu g/m^3$ (99th percentile) (Weisel et al., 2005), and

270 $\mu g/m^3$ (95th percentile) from a review of 12 residential studies (Logue et al., 2011). While individuals were not asked about their use of deodorizers and repellents, and the walk-through inspection could only confirm the use of these products in a few obvious cases (e.g., boxes of mothballs were occasionally observed), the sharp rise in the concentration distributions can be used to differentiate the residences where PDCB products are used.

The variance components analysis shows that the variability of indoor concentrations was due mostly to between home-to-home variation (44%), seasonal variation (23%), and city variation (22%) (Table 2). Measurement uncertainty was only 10%. This pattern held among three cities (YP, DB, and DT). In AA, variability was predominantly due to seasonal (53%) and then between-home differences (32%). The between-residence variation likely is attributable to differences in PDCB use patterns, source strengths, and ventilation among the homes, while the seasonal variation likely reflects changes in source strengths and ventilation. PDCB concentrations in bedrooms exceeded levels in living rooms in most (77%) of the DT homes (Wilcoxon pair-signed rank test, $P < 0.01$, $N = 279$), and the median concentration in the bedroom (0.72 $\mu g/m^3$) was 32% higher than that in the living room (0.52 $\mu g/m^3$). However, this difference is small compared to home-to-home, seasonal, city-to-city, and measurement variance. Most (87%) of the study homes had forced air heating/cooling systems, and the generally modest PDCB concentration differentials within the homes reflect the well-mixed conditions typically seen in US homes (Batterman et al., 2007; Dodson et al., 2008).

PDCB and naphthalene

As noted earlier, both PDCB and naphthalene are used as pest repellents and deodorizers. Based on monitoring in the same homes using the same methods, a subset of residences had elevated concentrations of naphthalene (Batterman et al., 2012a). Like the PDCB trend, the DT homes had the highest concentrations among the four cities, indicating that DT residents use repellents and deodorizers more frequently and more intensively than the families in the other communities. Indoor levels of the two chemicals, plotted in Figure 2, show moderate correlation (Spearman's correlation coefficient = 0.31, $P < 0.01$, two-tailed). A two-way contingency table in which each compound was grouped by quartiles showed 10.4% of residences in the top quartiles of both PDCB and naphthalene levels, and 12.5% in the bottom quartiles of the two chemicals, compared to 6.25% expected if use was uncorrelated (Table 3). Of the study residences, 3% had both PDCB and naphthalene above their 90th percentile concentrations (7.0 and 6.4 $\mu g/m^3$, respectively), compared to the 1% expected if use was uncorrelated.

In comparison with naphthalene, the distribution of PDCB concentrations was much more skewed. This is

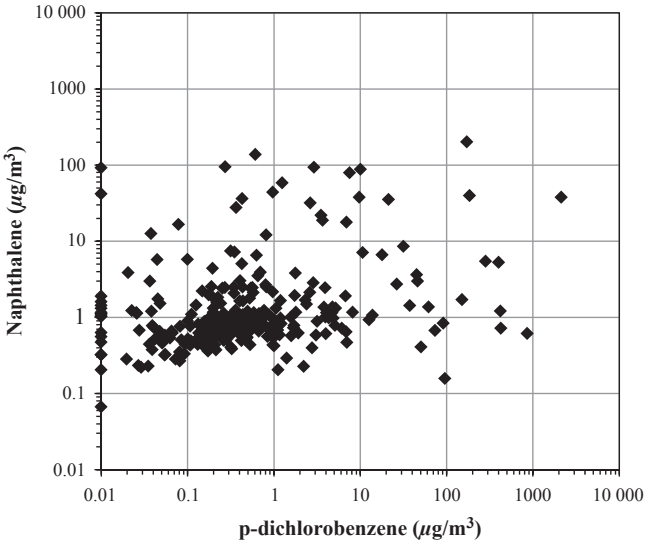

Fig. 2 Scatterplot of indoor *p*-dichlorobenzene and naphthalene concentrations ($N = 287$)

Table 2 Variance proportions (%) for indoor and outdoor *p*-dichlorobenzene concentrations

Location/component	AA	YP	DB	DT	All
Indoors					
City	–	–	–	–	22.3
Residence	32.1	76.4	62.9	61.3	44.2
Within-home	–	–	–	0.0	–
Season	52.6	14.3	25.9	26.9	23.4
Measurement	15.3	9.2	11.2	11.8	10.0
Outdoors					
City	–	–	–	–	15.7
Residence	0.0	44.4	1.3	–	14.1
Season	7.4	0.0	43.3	–	15.4
Measurement	92.6	55.6	55.4	–	54.7

AA, Ann Arbor; DB, Dearborn; DT, Detroit; YP, Ypsilanti.

Table 3 Joint frequency of *p*-dichlorobenzene and naphthalene concentrations across the four cities by concentration quartile (concentration ranges indicated)

Naphthalene		*p*-dichlorobenzene				
		<P25	P25–P50	P50–P75	>P75	
Percentile	Conc.($\mu g/m^3$)	<0.15	0.15–0.36	0.36–1.11	>1.11	Subtotal
<P25	<0.58	36	16	10	8	70
P25–P50	0.58–0.89	12	26	21	15	74
P50–P75	0.89–1.64	15	15	24	18	72
>P75	>1.64	11	12	18	30	71
	Subtotal	74	69	73	71	287

p-dichlorobenzene and naphthalene distributions were based on residence-based statistics for all four communities.

demonstrated by skewness coefficients of 8.2 for PDCB vs. 5.1 for naphthalene, the ratio between peak and median concentrations (19-fold for 90th percentile and median concentrations of PDCB and sevenfold for naphthalene), and peak concentrations (2100 $\mu g/m^3$ for the highest seasonal averaged PDCB concentration vs. 200 $\mu g/m^3$ for naphthalene). The top deciles of PDCB and naphthalene concentrations fit GEV distributions (Anderson–Darling test, $P = 0.86$ and 0.76, respectively). The greater skewness of PDCB concentrations likely results from several factors. As noted earlier, PDCB has no other sources besides moth and pest repellents, unlike naphthalene, which has many sources that cause it to be ubiquitous in air (Batterman et al., 2012a; Jia and Batterman, 2010). Second, only a subset of residences uses these products. Third, PDCB has a higher vapor pressure than naphthalene, that is, a comparably sized cake or ball will produce a higher concentration, albeit for a briefer period, and thus, monitoring may be more likely to encounter peak concentrations associated with recent use of PDCB.

Monitoring results show that many households in the four communities use both products and that DT residents use both chemicals most often. These products are marketed somewhat differently, for example, local stores offer 'moth balls' comprising either naphthalene or PDCB, while most toilet cakes or tablets contain PDCB. However, product labeling does not emphasize the product's composition, and we suspect few users know about the composition, and thus, the products are used interchangeably.

Outdoor concentrations

Outdoor PDCB levels at residences in AA, YP, and DB averaged only 0.09 $\mu g/m^3$ (median of 0.04 $\mu g/m^3$; $N = 145$; Table 1). Based on the variance proportions analysis, measurement variability contributed 55% of the total variance, a high level attributable to concentrations that frequently were near the MDL ($\sim$0.02 $\mu g/m^3$). Effects of city, season, and house-to-house variation were similar, each contributing 14–16% of the variance (Table 2). Variance analyses by city again showed that measurement uncertainty was largest (55–93%); contributions of seasonal and residence effects differed by city. The small between-house variance indicates that concentrations were relatively homogeneous within a neighborhood. Similar PDCB levels have been seen across industrial, residential, and commercial sites in DT (Simon et al., 2005) and Yokohama, Japan (Tiwari et al., 2010). Because outdoor samplers were located near the houses, some of our measurements may have reflected air exhausted from houses, or possibly the use or disposal of PDCB near the house.

The mean concentration outside homes in AA, YP, and DB was at the low-end of the ranges reported outside homes (0.1–5 $\mu g/m^3$), and the median was within the 0.1–1.8 $\mu g/m^3$ range reported outside homes (Aronson et al., 2007; Johnson et al., 2010; Sexton et al., 2004; Weisel et al., 2005). Measurements at fixed ambient sites show a slightly larger range, 0.18–1.2 $\mu g/m^3$, based on measurements in 11 other US states from 1998 to 2008 (Hultin et al., 2010; Simon et al., 2005). While PDCB concentrations outside of the DT homes were not obtained in the present study, levels at six DT sites in 2000–2001 ranged from 0.2 to 0.3 $\mu g/m^3$ and detection rates were only 16–35%; levels at a seventh site (N. Delray, a population-oriented site) averaged 3.5 $\mu g/m^3$ ($N = 29$) (Simon et al., 2005). In 2006–2007, PDCB levels at one of the DT sites (DB) decreased to 0.1 $\mu g/m^3$ from 0.2 $\mu g/m^3$ in 2000–2001 (Hultin et al., 2010), which was similar to our DB measurements.

The highest outdoor levels (integrated over 3–4 days) ranged from 0.14 (AA) to 1.7 $\mu g/m^3$ (YP). Much higher short-term levels have been reported: 83 $\mu g/m^3$ (24 h) in DT in 2001–2002 (Simon et al., 2005); 3.6 $\mu g/m^3$ (integrated 7 days) in DT in 2006 (Johnson et al., 2010); 355 $\mu g/m^3$ in New Jersey in 2000 (The Health Effects Institute, 2008); 1.0–120 $\mu g/m^3$ in relatively old studies reviewed by ADSTR (ATSDR, 2006); 8.4 $\mu g/m^3$ in West Louisville, Kentucky in 2000 (Simon et al., 2005); and 16 $\mu g/m^3$ in industrial sections of Toronto and Windsor, Canada (Environment Canada, 1993). These levels indicate local sources or, we suspect, contamination of the sample because PDCB is widely used indoors.

Outdoor PDCB concentrations were far lower than indoor levels in most homes. Thus, outdoor levels will provide only small contributions to the total exposure for most persons. For those homes that do not contain PDCB-emitting products, however, outdoor levels represent a 'floor' for indoor concentrations, and thus, indoor and outdoor levels will be very similar.

Sources and emission rates of *p*-dichlorobenzene

Emission rates of PDCB-based products have been estimated using both chamber and test house experiments. Emission rates of 3.3–11 mg/h were estimated for individual tablets (7.5–8.4 g each), 3.7 mg/h for a block-type product (26 g), and 350 mg/h for a hanging dispenser (160 g), all at 25°C using a model for products sold in Japan (Shinohara et al., 2008). Using small chamber tests and air exchange rates from 0.25 to 2.0 per h, emission rates from moth crystal cakes (surface area 55 cm^2 each) ranged from 58 to 99 mg/h at 23°C and 220 to 325 mg/h at 35°C (Tichenor et al., 1990). Five moth crystal cakes placed in the closet of a test house gave PDCB emission rates from 381 to 464 mg/h over an 11-day period (Aronson et al., 2007). Emission rates for toilet bowl deodorizers (85 g

toilet rim block) reported by the US Consumer Products Safety Commission in 1991 averaged 310 and 588 mg/h in 13- and 5.5-day-long tests, respectively (Aronson et al., 2007). Several other studies have reported concentrations (but not emission rates) from PDCB deodorizers: Median and average personal air concentrations were 340 and 500 $\mu g/m^3$ in 3-day tests of toilet bowl deodorizers (Wallace et al., 1989); concentrations reached 871 $\mu g/m^3$ in residential bathrooms containing deodorizer blocks (Djohan et al., 2007); and spray and liquid deodorizers increased indoor air concentrations to 37 and 25 $\mu g/m^3$, respectively (Wallace et al., 1989). Additionally, six of 26 gel-type air fresheners tested in Korea contained PDCB, but emission rate information was not presented (Jo et al., 2008).

Based on Equation 1 and the nominal building parameters, the 90th percentile concentration in DT residences (26 $\mu g/m^3$) yields an emission rate of 5.3 mg/h, which approximately matches emission rates for a single moth 'ball' or tablet. For the maximum concentration (2100 $\mu g/m^3$), the emission rate prediction is 431 mg/h, which is equivalent to that from one or two block-type products or boxes of moth 'crystal cakes'. Of course, residences can contain multiple PDCB sources, and the emission rate estimates can be affected by mixing assumptions, the presence of exhaust ventilation, adsorption/desorption (source–sink) effects, and the location of the source, for example, bathroom, closet, and garment bag. Emission rates also may slowly decline as the mass of the PDCB solid decreases (Shinohara et al., 2008).

The emission rates derived from the DT data, which represent real-world conditions, appear reasonable based on the available literature. Emission test results depend on test conditions, for example, product type, amount, temperature and air exchange rate, and in-use conditions may involve mass-transfer limitations that can significantly lower emission rates compared to those determined using chamber tests. This may be especially important for moth repellents, which are often used in closed spaces with limited airflow, for example, garment bags, drawers, and closets. Use of PDCB-containing products in homes in our study is suggested by PDCB concentrations exceeding 1–2 $\mu g/m^3$, a level that corresponds to a rapid increase in the PDCB concentration distribution (Figure 1). This threshold suggests PDCB use in approximately 15% (AA) to 35% (DT) of the study homes.

Health risks

Long-term concentrations of PDCB were far below the current chronic non-cancer RfC (800 $\mu g/m^3$) in nearly all homes (100% of AA, DB, and YP homes, 96% of DT homes). Distributions of lifetime excess cancer risks in the four cities predicted using the California URE (1.1×10^{-5} per $\mu g/m^3$) are shown in Figure 1 on the right-hand axis. For the median and 90th percentile PDCB concentrations across the four cities, the predicted risks are 4.0×10^{-6} and 7.7×10^{-5}, respectively. Depending on the city, 3–13% of homes exceeded 9.1 $\mu g/m^3$, a risk level of 10^{-4}, 0–6% of homes exceeded 91 $\mu g/m^3$, a risk level of 10^{-3}, and 1% of homes exceeded 910 $\mu g/m^3$, a high risk of 10^{-2}. Five homes in DT (4%) exceeded 240 $\mu g/m^3$, the Japanese indoor air guideline. Similar results have been shown in several studies, for example, median risks of PDCB ranged from 10^{-4} to 10^{-6} (Loh et al., 2007), and 90th percentile risks for PDCB exceeded 10^{-3} in the RIOPA study (Hun et al., 2009) and in some high schools in New York City and Los Angeles (Sax et al., 2006).

Estimated cancer risks from PDCB at high concentrations exceed those for most other VOCs, for example, benzene (Hun et al., 2009; Jia et al., 2008b), as well as other pollutant types with the possible exceptions of particulate matter and radon. However, PDCB risks are about an order of magnitude below those from naphthalene [median and 90th percentile risks of 9×10^{-5} and 7×10^{-4}, respectively, calculated using the draft inhalation URE of 1×10^{-4} per $\mu g/m^3$ under consideration by US EPA (2004) Batterman et al., 2012a].

Discussion

Significance

This is the first paper that examines concentrations, exposures, and risks of PDCB in indoor and outdoor air in Michigan, and the concentration distributions and variance components analyses demonstrate that both the frequency of PDCB use and usage practices differed among the four cities tested. In particular, PDCB was seen more frequently at higher concentrations in DT homes than in the other cities. Of the four communities, DT has the lowest household income, lowest educational attainment, poorest health by many indicators, and the highest fraction of African Americans. Discussions with community members indicate that deodorants and repellants (including PDCB and naphthalene) are sometimes used in an 'off-label' and inappropriate manner, for example, to mask odors from pesticide applications. Potentially, the poorer quality of housing that leads to greater frequency of odor and pest issues, and attitudes and behaviors that lead to greater acceptance of chemical use in homes are key reasons why DT showed the most frequent use and the highest concentrations of PDCB across the four communities. This suggests a need to target policies and tailor educational and awareness efforts to the groups that are most at risk.

The study is also significant in showing that high concentrations of PDCB are not uncommon, the highly skewed nature of the concentration, and the low temporal correlation of concentrations in the households, that is, a high concentration in a home in one season often was not seen in subsequent seasons. This low correlation, along with the variance components analysis, indicates that repeated measurements of PDCB in a large number of homes are needed to estimate long-term averages and exposures that are representative.

Risks and risk management

p-dichlorobenzene's very skewed concentration distribution produces much higher risks for a subset of residences, for example, risks exceeded 10^{-3} for 4% of homes, indicating that PDCB exposure can be significant. The current strategy for managing exposures and risks associated with most consumer and industrial products is through right-to-know requirements. PDCB is listed as a carcinogen in California's Proposition 65, in Michigan's air toxics guidance, in the Safe Drinking Water and Toxic Enforcement Act of 1986, and as a hazardous chemical under the European Union's Regulation on Registration, as examples. In the Registration Eligibility Decision for PDCB, US EPA initially specified a draft inhalation cancer unit risk of 4×10^{-6} per $\mu g/m^3$, but this was withdrawn because of the lack of evidence regarding carcinogenicity (USEPA, 2008). US EPA then classified PDCB as a low-risk pesticide (USEPA, 2010). Both PDCB and naphthalene are commonly used as deodorizers and pest repellents in residential and other settings. Sometimes these products are applied as insect and animal repellents by placing flakes or tablets on trays or other surfaces in rooms, attics, and outdoors in gardens. Such excessive or 'off-label' uses can greatly elevate indoor concentrations. While naphthalene has lower sublimation rates and thus lasts longer, the US market has shifted from naphthalene to PDCB owing to its lower toxicity. At the same time, fragrances, essential oils, and surfactant-based alternatives have replaced both PDCB and naphthalene as air freshener and toilet/urinal deodorizers. California has restricted the sale of consumer products containing PDCB, including air fresheners and toilet/urinal care products (California Air Resources Board, 2003; New York State, 2004), and New York has restricted the purchase and use of PDCB-containing toilet or urinal deodorizers in school buildings (New York State, 2004). Still, PDCB is widely available and use remains high. The high concentrations found in the present study demonstrate a need to manage PDCB exposures and risks. Possible actions might include sales restrictions, improved labeling and use instructions, consumer education, and promotion of non-toxic alternatives.

Study limitations

This study has several limitations. With respect to the experimental measurements, while homes were measured at least twice, seasonal variability was only partially captured. In AA, YP, and DB homes, indoor samples included only the living room, and concentrations in other indoor locations (bathrooms, basement, garage, vehicles, workplaces, etc.) where people might be exposed were not measured. As mentioned, we did not document or ask about specific uses and application rates of PDCB in each home, and thus, we cannot determine whether mothballs, room deodorizers, toilet deodorizers, or some other source is responsible for the high concentrations found. The study households may not be representative of those in Michigan or the United States more broadly. While several sources associated with high PDCB concentrations were identified, some of the analyses assumed building characteristics and used PDCB emission rates from the literature. Lastly, the health risks presented are screening level estimates. They were calculated using indoor air measurements, which do not represent lifetime exposures, and they may be biased upwards because they do not account for the amount of time that individuals spend in homes, temporal variation, other exposure sources, and other microenvironments.

Conclusions

p-dichlorobenzene is widely used indoors as a pest repellant and deodorizer. Long-term average concentrations of PDCB measured in four Michigan cities ranged from 0.2 to 1.7 $\mu g/m^3$, similar to the levels found in earlier studies. Concentration distributions were strongly right-skewed, and greatly elevated levels were seen in a subset of homes. DT homes showed the highest concentrations and the highest use of PDCB. In this city, 4% of study homes exceeded 800 $\mu g/m^3$, the RfC for non-cancer effects, and the highest excess individual cancer risk estimates exceeded 10^{-3}. House-to-house variation was large, reflecting differences in PDCB use between residences. These results, in particular the number of homes that had excessive concentrations of PDCB, suggest a need to eliminate or restrict the use practices of this chemical.

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