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An Investigation of Homes with High Concentrations of PCDDs, PCDFs, and/or Dioxin-Like PCBs in House Dust

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*As part of the University of Michigan Dioxin Exposure Study, the 29 congeners of polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans, and dioxin-like polychlorinated biphenyls that have World Health Organization consensus toxic equivalency factors were measured in house dust from 764 homes using a population-based sampling design over selected regions in five Michigan counties. Twenty homes had a total toxic equivalency in house dust that was more than 2.5 standard deviations above the mean (i.e., defined to be outliers). This follow-up investigation describes the outlier house dust measurements and corresponding soil measurements and explores possible sources of these toxins in house dust. The congener distributions in the house dust outliers varied and were dominated (i.e., >50% of the total toxic equivalency) by either polychlorinated dibenzo-*p*-dioxins (n = 9), polychlorinated dibenzofurans (n = 1), or dioxin-like polychlorinated biphenyls (n = 9). Likely sources of contamination of house dust were identified in only three cases. In two cases, dust contamination appeared to be related to contaminated soil adjacent to the home; in one case, contamination was related to a source within the home (a carpet pad). In most cases, the source(s) of contamination of house dust could not be identified but appeared likely to be related to uncharacterized sources within the homes.*

[Supplementary materials are available for this article. Go to the publisher's online edition of the Journal of Occupational and Environmental Hygiene *for the following free supplemental resource: a PDF file containing a summary of baseline and dust outlier interview questions and tables containing PCB, PCDD, and PCDF concentrations in various samples of house dust and soil.]*

Keywords dioxins, furans, house dust, polychlorinated biphenyls

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INTRODUCTION

Few studies have measured polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and/or dioxin-like polychlorinated biphenyls (PCBs) in house dust by vacuum sampling, and most of those studies have been small (fewer than 25 houses sampled).[1–4] No previous study has obtained and analyzed vacuum house dust samples for PCDDs, PCDFs, and dioxin-like PCBs from a large, representative sample of homes, most of which were not suspected of contamination.

The University of Michigan Dioxin Exposure Study (UMDES) was designed to determine whether PCDDs, PCDFs, and dioxin-like PCBs (hereafter collectively referred to as dioxins) in soil and/or house dust are related to or explain serum levels of these contaminants, with adjustment for other known risk factors (i.e., age, sex, diet, occupation, body mass index, smoking, breast feeding history, etc.).

The study was undertaken in response to concerns among the population of Midland and Saginaw counties that dioxin-like compounds from the Dow Chemical Company facilities in Midland, Michigan, have contaminated areas of the City of Midland and sediments in the Tittabawassee River floodplain.[5] The study measured the levels of the World Health Organization (WHO) 29 dioxin congeners with consensus toxic equivalency factors (TEFs) in serum, soil, and house dust from a population-based sample of residents in the study regions.[6] Not surprisingly, the distribution of total toxic equivalencies (TEQs) in house dust was skewed (roughly log-normal), with a small number of high or outlier values. If house dust is an important pathway of exposure to dioxins, then these outlier cases would be of most interest, and it is important to better understand the sources of such contamination. This

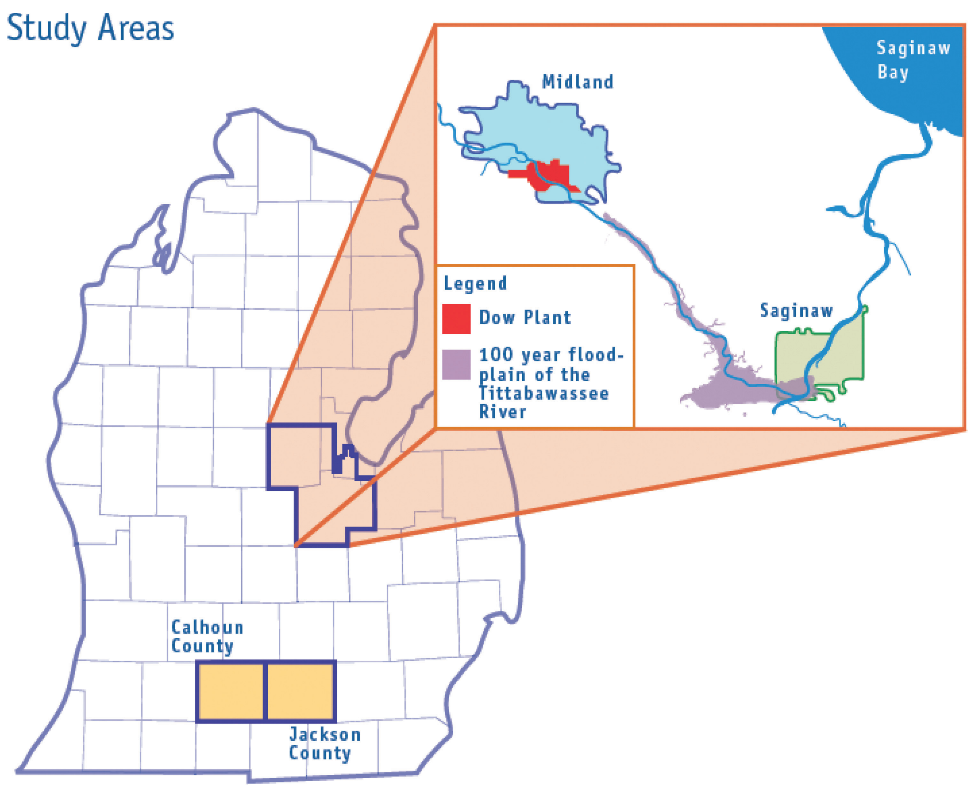

FIGURE 1. Study areas in state of Michigan

follow-up investigation describes the outlier house dust measurements and corresponding soil measurements and explores possible sources for high TEQ values in house dust.

METHODS

The UMDES involved a two-stage clustered random sampling design to recruit subjects from five regions in the state of Michigan. The regions were: the 100-year Federal Emergency Management Agency (FEMA) floodplain of the Tittabawassee River or whether the respondent reported flooding of the home by the Tittabawassee River (FP); the near-floodplain of the Tittabawassee River (NFP); the plume area in the City of Midland downwind from the historic incineration activities of the Dow plant (PL); elsewhere in Midland and Saginaw counties and parts of Bay County outside the floodplain, near-floodplain, and plume areas (MS); and Jackson and Calhoun counties (located more than 200 kilometers away from the Dow facilities in Midland) that served as a control area (JC, Figures 1 and 2). Eligible subjects were required to be at least 18 years old and to have lived in their homes for at least 5 years. Data collection for the main study was completed in 2004–2005, and involved an hour-long interview and obtaining blood, house dust, and soil samples for chemical analyses from eligible subjects.

Subjects who were interviewed ($n = 1324$) needed to meet additional criteria to be eligible to provide blood, soil and/or dust samples (see Figure 3 for a summary of participation at each stage of recruitment). Recruitment percentages did not differ among the study regions, and so only aggregate numbers are shown in Table I and Figure 3. Vacuum house dust samples were obtained from 764 homes: 207 from the floodplain of the Tittabawassee River, 159 from the near-floodplain of the Tittabawassee River, 37 from the Midland plume area, 163 from elsewhere in Midland and Saginaw counties, and 198 from Jackson and Calhoun counties (Table I). Soil samples were obtained from 736 of the 764 homes from which house dust samples were obtained.

A dust outlier result was defined as one with a total TEQ (picograms per gram of house dust or parts per trillion, ppt) that was more than 2.5 standard deviations (SD) above the mean of the log-transformed data. Note that this investigation was initiated in mid-2005, and the determination of outlier status was based on calculation of the TEQs using the 1998 TEFs, but all TEQ results shown here have been recalculated using the 2005 TEFs.[7,8]

Twenty house dust results (2.6%) met the statistical criterion to be classified as outliers (see Table II and online supplemental Table II for listing of dust outlier results). If the 2005 TEFs had been used instead of the 1998 TEFs, 15 of the 20 original cases would have met the criterion for being classified as an outlier (i.e., a TEQ more than 2.5 SD above the mean of the log-transformed data), but no new cases met the criterion. All 20 cases are presented in this report.

TABLE I. Number of Study Participants and Samples

	Floodplain (FP)	Near Floodplain (NFP)	Midland/ Plume (PL)	Other Midland/ Saginaw (MS)	Jackson/ Calhoun (JC)	Total All Areas
Interviews	326	264	71	304	359	1324
Blood samples	251	197	48	199	251	946
House dust samples	207	159	37	163	198	764
Soil samples	203	164	37	168	194	766
Interviews, blood, dust, and soil samples	195	156	35	162	183	731

Subjects were given the option to receive the results of the chemical analyses of their house dust, and overall, 67% of subjects elected to receive their results. Only subjects who opted to receive these results were eligible to participate in the follow-up investigation. Thirteen of the 20 outlier subjects had elected to receive results and were invited to be reinterviewed as part of this house dust follow-up study. Follow-up interviews were completed with all 13 eligible subjects (Table III).

The follow-up interview included questions covering the following topic areas: house characteristics (e.g., age of house, age of carpets/rugs, presence/use of a wood-burning fireplace, trash burning, flooding from the Tittabawassee River, remediation, professional cleaning of floors/rugs/carpets, use of pesticides inside the house, history of renovations or structural fires, nearby industries); characteristics/habits of occupants of the house (e.g., ages of occupants/presence of children, pets,

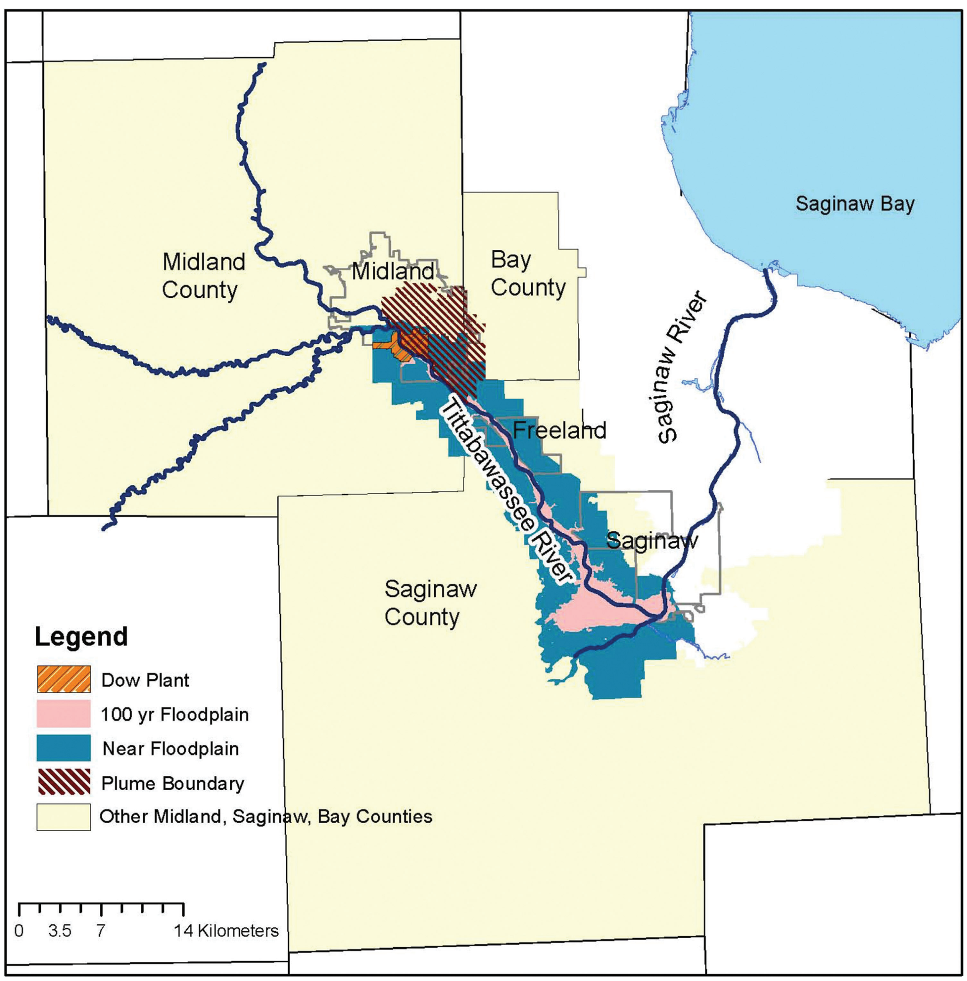

FIGURE 2. Detail of study areas in Midland, Saginaw, and Bay counties, Michigan

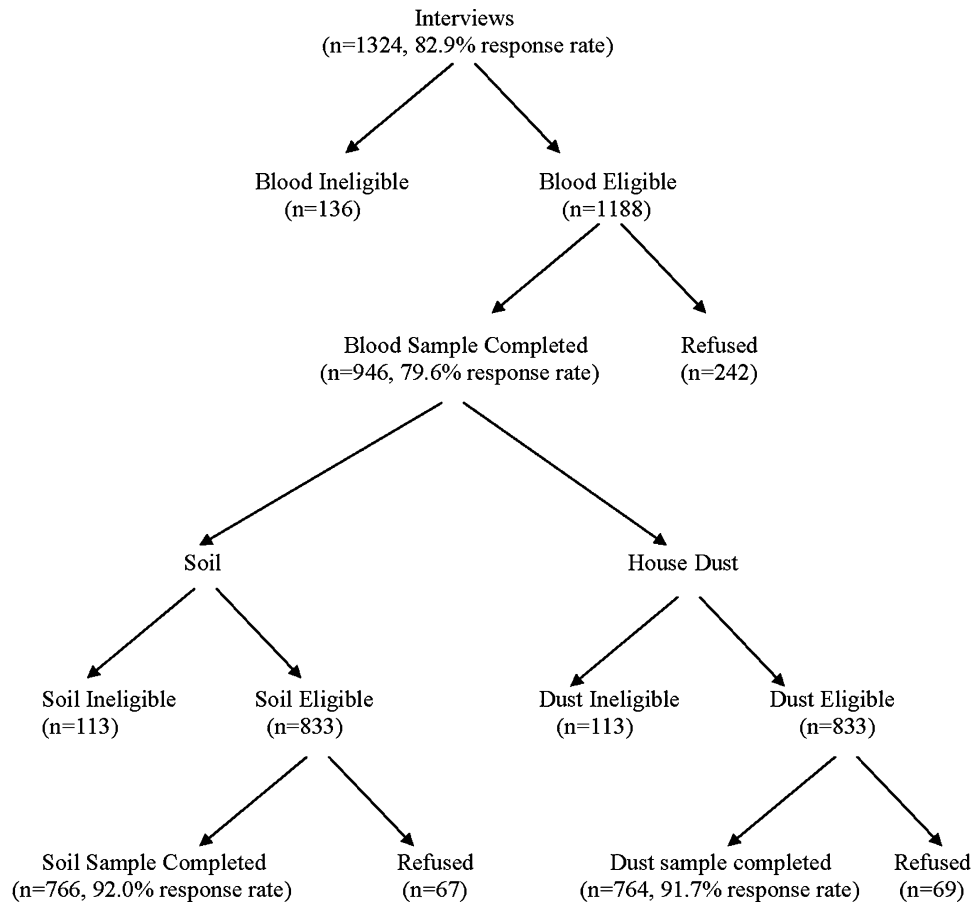

FIGURE 3. Recruitment of subjects

smoking habits, occupations and hobbies of all residents, gardening activities, shoe removal); and other items of potential interest (e.g., treated wood in/around house, potted plants in house) (see supplemental Table I for a partial summary of follow-up interview questions). Some of these topics were also covered in the main study interview, and so limited information was available for subjects who were not eligible for the follow-up interview.

House dust vacuum samples were collected using High Volume Small Surface Samplers (HVS3s) manufactured by CS-3, Inc. (Sandpoint, Idaho). Vacuums were equipped with a cyclone and fine particle filter capable of capturing 99.95% of particles above 0.3 micrometer aerodynamic mean diameter. One composite sample, typically based on vacuuming from two separate locations, was taken in each household from sampling locations that presented the highest potential for human contact with dust. Locations were generally a frequently occupied living space (e.g., the middle of a family room) and a high traffic pathway (e.g., a main hallway). Samples were taken from both hard and soft surfaces, with carpets and area rugs being preferred sampling surfaces. Samples were not taken of undisturbed dust in generally inaccessible areas. The sampling protocol was based, with minor modifications, on the ASTM method Standard Practice for Collection of Floor Dust for Chemical Analysis.[9] The sampling technicians attempted to collect a minimum of 10 grams of total dust. Samples were transported on wet ice to a dedicated 4°C cooler until delivery to the laboratory. Analyses of the samples was performed for the 29 dioxin-like congeners with WHO TEFs using internal modifications of the U.S. Environmental Protection Agency (USEPA) methods 8290 and 1668.[10,11] Results below the limit of detection (LOD) are reported as the LOD/$\sqrt{2}$.[12]

As noted above, in many cases dust results for the main study were based on composite samples, and so a high result may have been due to contamination in only one location from which the composite sample was derived. As part of the follow-up investigation, repeat dust samples were sought from the exact same locations in each home using the same equipment, sampling procedures, laboratory, and laboratory methods, except that the repeat dust samples were not composited. This provided the opportunity for more precise localization of dust contamination within each home. Repeat sampling and analyses of house dust were completed in 12 of the 13 homes that were eligible (supplemental Table V).

TABLE II. Soil and Baseline House Dust Test Results for Subjects with Outlier House Dust Results

Case Number Location[A]	1 FP	2 JC	3 JC	4 JC	5 JC	6 MS	7 MS	8 JC	9 JC	10 PL	11 JC	12 MS	13 FP	14 FP	15 JC	16 MS	17 MS	18 JC	19 MS	20 FP
Reinterviewed?[B]	Yes	Yes	No	Yes	Yes	Yes	No	No	No	Yes	No	Yes	Yes	No	Yes	No	Yes	Yes	Yes	Yes
Baseline Dust %TEQ from PCDDs	2.6	26.2	14.8	3.4	8.8	41.5	94.3	95.1	3.8	4.2	12.2	88.5	12.5	91.3	93.2	88.9	49.3	93.6	93.4	98.7
Baseline Dust %TEQ from PCDFs	22.6	16.7	3.2	5.2	5.0	7.4	4.7	4.2	10.0	9.8	4.6	6.6	84.8	5.0	6.6	10.4	15.9	4.4	6.4	1.2
Baseline Dust %TEQ from PCBs	74.8	57.1	82.0	91.3	86.2	51.1	1.1	0.7	86.2	86.1	83.2	4.9	2.7	3.7	0.2	0.8	34.8	2.0	0.2	0.1
Baseline Dust Total TEQ (ppt)	150	176	197	199	224	268	283	301	323	334	382	408	411	497	544	566	824	1110	1400	1750
Maximum Soil %TEQ from PCDDs[C]	57.3	42.8	81.5	50.2	2.14	79.5	19.2	83.4	23.9	70.4	33.7	41.6	NA	60.2	23.5	58.2	21.6	70.81	31.48	NA
Maximum Soil %TEQ from PCDFs	37.9	19.7	7.56	26.2	3.22	18.2	78.6	12.4	49.3	26.6	17.7	44.5	NA	25.2	26.8	31.7	57.4	21.30	51.53	NA
Maximum Soil %TEQ from PCBs	4.78	37.5	11.0	23.6	94.6	2.37	2.15	4.14	26.8	2.92	48.6	13.9	NA	14.6	49.7	10.1	21.0	7.90	17.00	NA
Maximum Soil Total TEQ (ppt)	31.8	13.9	6.35	7.59	72.8	8.71	21.5	3.68	10.8	60.6	2.59	5.27	NA	4.15	6.92	7.20	24.0	19.1	16.6	NA
House Perimeter Soil %TEQ from PCDDs	54.7	42.8	81.5	50.2	17.0	79.5	25.2	59.9	31.9	74.6	39.4	41.6	NA	60.2	30.7	58.2	34.8	70.81	27.32	NA
House Perimeter Soil %TEQ from PCDFs	16.5	19.7	7.56	26.2	13.3	18.2	71.0	33.1	34.8	23.1	16.4	44.5	NA	25.2	30.3	31.7	40.9	21.30	60.28	NA
House Perimeter Soil %TEQ from PCBs	28.8	37.5	11.0	23.6	69.8	2.37	3.81	6.96	33.3	2.23	44.2	13.9	NA	14.6	39.0	10.1	24.3	7.90	12.40	NA
House Perimeter Soil Total TEQ (ppt)	15.0	13.9	6.35	7.59	7.90	8.71	13.2	1.83	10.52	52.7	2.29	5.27	NA	4.15	5.28	7.20	19.1	19.12	15.43	NA

[A] The floodplain of the Tittabawassee River (FP), the near-floodplain of the Tittabawassee River (NFP), the Midland plume area downwind from the historic incineration activities of the Dow plant (PL), elsewhere in Midland and Saginaw counties (MS), and Jackson and Calhoun counties that served as a control area (JC).

[B] See supplemental Table I for details of interview questions.

[C] TEQ calculated based on WHO 2005 TEFs. All TEQ results are reported as parts per trillion (ppt) on a dry weight basis.

NA, not available/not obtained.

TABLE III. Summary of Interviews of Subjects with Outlier House Dust Results

Case Number Location[A]	1 FP	2 JC	3 JC	4 JC	5 JC	6 MS	7 MS	8 JC	9 JC	10 PL
Reinterviewed?[B]	Yes	Yes	No	Yes	Yes	Yes	No	No	No	Yes
Year house was built[C]	1960–65	1925–30	NA	1920–25	1960–65	1965–70	NA	NA	NA	1915–20
Duration of occupancy (years)[C]	30–35	45–50	55–60	20–25	30–35	15–20	25–30	20–25	5–10	10–15
Age carpet/rug/flooring (years)[C]	10–15	10–12	10	15	20–40	3–6	20	17	>50	17–22
Fireplace(s)?	Yes	Yes	No	No	Yes	Yes	No	Yes	Yes	No
Flooding from TR?	No	No	No	No	No	No	No	No	No	No
Indoor/outdoor pet(s)?	Yes	No	No	Yes	No	Yes	Yes	No	Yes	Yes
Relevant occupation(s)?	Yes	No	No[F]	No	No	Yes	NA[F]	No[F]	No[F]	No
Relevant hobbies?	No	No	NA	No	No	No	NA	NA	NA	No
Regularly remove shoes?	Yes	No	No	No	No	No	No	No	No	No
Pesticide sprayed inside home?	No	No	NA	No	No	No	NA	NA	NA	No
Professional carpet cleaning?	No	No	NA	Semiannual	No	No	NA	NA	NA	Annual
Fire damage to home?	No	No	No	No	No	No	No	No	No	No
Waste burning?	No	Yes	No	No	No	No	No	Yes	Yes	No
Regular smoking in the home?	Yes	Yes	No[F]	Yes	Yes[G]	No	Yes	No[F]	Yes	Yes
Children under 18?	Yes	Yes	Yes	Yes	No	Yes	Yes	Yes	NA	Yes

Case Number Location[A]	11 JC	12 MS	13 FP	14 FP	15 JC	16 MS	17 MS	18 JC	19 MS	20 FP
Reinterviewed?[B]	No	Yes	Yes	No	Yes	No	Yes	Yes	Yes	Yes
Year house was built[C]	NA	1955–60	1970–75	NA	1945–50	NA	<1910	<1910	1945–50	1997–99
Duration of occupancy (years)[C]	40–45	40–45	15–20	15–20	5–10	15–20	20–25	20–25	40–45	6–8
Age carpet/rug/flooring (years)[C]	6	10–15	5–7	>15	<5	4–15	>20	20–40	10–20	6–8
Fireplace(s)?	Yes	No	No	No	No	No	No	Yes[D]	No	No
Flooding from TR?	No	No	Yes	No	No	No	No	No	No	No
Indoor/outdoor pet(s)?	No[E]	Yes	No	No	Yes	Yes	Yes	Yes	Yes	No
Relevant occupation(s)?	No[F]	No	No	No[F]	No	No[F]	No	No	No	No
Relevant hobbies?	NA	No	No	NA	No	NA	No	No	No	No
Regularly remove shoes?	No	No	Yes	Yes	NA	No	No	No	No	No
Pesticide sprayed inside home?	NA	No	No	NA	No	NA	No	No	No	No
Professional carpet cleaning?	NA	~ 1993	~ 2005	NA	Annual	NA	No	No	~ 2004	No
Fire damage to home?	No	No	Yes	No	No	No	No	Yes	No	No
Waste burning?	Yes	No	No	No	No	No	No	No	No	No
Regular smoking in the home?	Yes	Yes	No	No[F]	No	No[F]	Yes	Yes	Yes	No
Children under 18?	NA	Yes	No	NA	Yes	NA	Yes	NA	No	No

[A] The floodplain of the Tittabawassee River (FP), the near-floodplain of the Tittabawassee River (NFP), the Midland plume area downwind from the historic incineration activities of the Dow plant (PL), elsewhere in Midland and Saginaw counties (MS), and Jackson and Calhoun counties that served as a control area (JC).
[B] See supplemental Table I for details of interview questions.
[C] Some answers are approximate to protect the confidentiality of respondents.
[D] Two woodburning fireplaces in the building, but not used since 1965.
[G] Pipe only; no cigarettes.
[F] Respondent only; no information on other members of household.
[E] None since 1985.
NA, not available/not obtained.

Soil sampling was conducted following the consent of the respondent. Properties were sampled in multiple locations using a push core sampler that collected a core of soil from the surface to 6 in. depth. Surface vegetation at the site of the core was also collected except in situations where garden plants might be damaged. Selection of locations for sampling followed a protocol that identified the house perimeter, property areas where skin contact was likely (e.g., up to two separate garden locations), and areas in or near the floodplain of the Tittabawassee River. Up to four stations were located around the four sides of the house, and a station was defined by laying out a 3 ft diameter ring. Three equally spaced 6-in. cores were collected from the interior of each ring (i.e., a total of 12 cores taken from the house perimeter in most cases). Cores were separated into 0–1 in. and 1–6 in. strata. Strata were composited across cores, resulting in a single 0–1 sample and a single 1–6 sample for chemical analyses from the house perimeter.[13]

The soil samples were archived in 100-g dioxin-grade, amber glass containers to avoid photolytic degradation reactions and stored in dedicated 4°C cold rooms prior to analysis. Analyses for the 29 dioxin, furan, and PCB congeners were performed by the same laboratory and using the same methods as for house dust. Soil samples were obtained from 18 of the 20 properties of dust outliers (Table II and supplemental Tables III and IV). No additional soil samples were sought as part of the follow-up investigation.

In five cases, other materials aside from house dust were sampled during the follow-up investigation and sent to the same laboratory for the same chemical analyses in an attempt to identify possible sources of house dust contamination. These items included: cigarette ash, wood from a beam that had been charred in a structural fire, a piece of carpet pad, wood from an outdoor deck, and a piece of leather from a leather couch (Table IV).

The original UMDES study and the follow-up study were reviewed and approved by the University of Michigan Health IRB.

RESULTS

PCDD, PCDF, and/or PCB concentrations were measured above the detection limits in all 764 house dust samples. The mean and median TEQ values for the 764 house dust measurements were 36.7 ppt and 16.2 ppt, respectively, and the 95th percentile was 126 ppt (Table V). The cut-point for determining outlier status (i.e., 2.5 SD above the mean) was 230 ppt based on 2005 TEFs.

Results of chemical analyses of house dust for the 20 outlier cases are summarized in Table II, which shows the total TEQ and percentage contribution to the TEQ from PCDDs, PCDFs, and PCBs for each case (the complete congener-specific baseline house dust results are shown in supplemental Table II). As noted above, seven subjects elected not to receive their results of house dust analyses; the 13 cases that received their results and were reinterviewed are marked in the table. Dust outliers were found in all study regions, including one house from the Midland Plume and nine houses from the control area and Jackson and Calhoun counties.

It is interesting to note that the Midland Plume region, the region likely to have had historical aerosol deposition of dioxins emitted from the Dow facilities, had the highest median concentration of dioxins in house dust (Table V). The congener distributions among the dust outliers varied considerably, with 50% or more of the TEQ of dust outlier samples due to PCDDs (n = 9), PCDFs (n = 1), and PCBs (n = 9). Note that dust from Case 17 did not have >50% of the TEQ attributable to any congener group.

Tracking of adjacent soil into homes was considered to be a possible external source of contamination of house dust among outlier cases. PCDDs, PCDFs, and/or PCBs were detectable in soil above the limits of detection in all 766 properties from which soil samples were obtained. The mean, median, and 95th percentile for house perimeter soil results in the control area (Jackson/Calhoun counties) were: 6.9 ppt, 3.6 ppt, and 22.6 ppt, respectively.

Soil samples were obtained for 18 of the 20 dust outlier cases, and the results are summarized in Table II, which shows the total TEQ and percentage contribution to the TEQ from PCDDs, PCDFs, and PCBs for the top 1 in. house perimeter sample and the soil sample with the highest TEQ from each property. The full congener summaries of soil results are shown in supplemental Tables III and IV. Among the 18 cases with available soil results, only Case 10 had a house perimeter soil result that was elevated above the 95th percentile for Jackson and Calhoun, but the congener distribution of the house perimeter soil (74.6% of TEQ from PCDDs) was not consistent with the house dust (86.1% of TEQ from PCBs); no other soil sample in this case appeared to be a match, and so it is unlikely that soil adjacent to the home was the source of house dust contamination in this case.

In Case 5, the TEQ of the house perimeter soil was not elevated (TEQ = 7.9 ppt; Table II), but the TEQ in the highest soil sample (obtained from a garden located near the front entrance to the home) was elevated (TEQ = 72.8 ppt; Table II). The congener distribution in the latter soil sample (94.6% of TEQ from PCBs) appeared to be reasonably consistent with the distribution in the corresponding house dust (86.2% of TEQ from PCBs). The home is located in Jackson/Calhoun and was built in the early 1960s (Table III). There are no children and no indoor/outdoor pets.

The current occupants have maintained a flower garden just outside the front door of the house. For many years they have added to the garden multiple bags of top soil purchased from garden centers in local "box" store retailers. No samples of the purchased top soil were available for analyses. It would appear that the garden soil is a likely source of contamination of the house dust, and it is possible that the garden soil may have been contaminated from added top soil purchased in local garden centers.

Although no soil sample was obtained from Case 13, it appears reasonable to infer that soil was the source of

TABLE IV. Concentrations (in ppt) of PCDDs, PCDFs, and PCBs in Other Household Samples

Case Number Location[A] Sample Type	12 MS Cigarette Ashes	17 MS Carpet Pad	18 JC Charred Wood	19 MS Wood from Deck	20 FP Leather from Couch
2,3,7,8-TCDD	2.00[B]	3.40	2.84[B]	4.68[B]	14.4[B]
1,2,3,7,8-PeCDD	2.07[B]	5.26	2.70[B]	4.42[B]	17.8[B]
1,2,3,4,7,8-HxCDD	4.75[B]	38.4	2.79[B]	9.26[B]	25.0[B]
1,2,3,6,7,8-HxCDD	4.72[B]	1810	3.49[B]	8.49[B]	28.9[B]
1,2,3,7,8,9-HxCDD	6.82[B]	616	3.53[B]	8.49[B]	26.4[B]
1,2,3,4,6,7,8-HpCDD	31.3	72300	47.4	32.9	39.5[B]
OCDD	637	334000	160	258	87.9
2,3,7,8-TCDF	1.35[B]	88.1	7.32	4.29[B]	23.4[B]
1,2,3,7,8-PeCDF	1.82[B]	65.1	3.63	3.40[B]	13.6[B]
2,3,4,7,8-PeCDF	1.71[B]	374	9.58	3.15[B]	13.7[B]
1,2,3,4,7,8-HxCDF	0.88[B]	810	7.78	2.93[B]	16.1[B]
1,2,3,6,7,8-HxCDF	0.83[B]	299	5.35	2.91[B]	15.4[B]
1,2,3,7,8,9-HxCDF	1.34[B]	74.7	2.45[B]	3.10[B]	22.7[B]
2,3,4,6,7,8-HxCDF	1.00[B]	103	5.87	2.09[B]	17.5[B]
1,2,3,4,6,7,8-HpCDF	1.36[B]	565	20.7	4.65[B]	18.3[B]
1,2,3,4,7,8,9-HpCDF	1.51[B]	86.1	2.08	2.98[B]	17.9[B]
OCDF	4.42[B]	1140	12.4	9.05[B]	25.7[B]
PCB 81	2.13[B]	2170	65.5	3.03[B]	19.8[B]
PCB 77	7.23	68000	2020	43.7	123
PCB 126	3.92[B]	9900	45.2	9.62[B]	54.0[B]
PCB 169	2.00[B]	74.70	1.94	3.55[B]	14.1[B]
PCB 105	53.2	13800000	11300	1620	2020
PCB 114	6.41[B]	855000	487	29.5[B]	138[B]
PCB 118	163	33600000	23900	3560	6000
PCB 123	6.34[B]	439000	403	48.7	278[B]
PCB 156	8.15	3170000	1960	266	1620
PCB 157	1.92[B]	533000	402	49.7	229
PCB 167	4.42	1300000	637	91.9	659
PCB 189	1.44[B]	43600	68.6	6.52[B]	228
Total TEQ (ppt)[C]	7.81	3950	19.1	16.0	61.3
PCDD Contribution (%)	79.5	27.3	36.9	77.7	66.4
PCDF Contribution (%)	14.6	6.55	31.9	16.6	23.5
PCB Contribution (%)	5.91	66.2	31.3	7.76	10.1

[A] The floodplain of the Tittabawassee River (FP), elsewhere in Midland and Saginaw counties (MS), and Jackson and Calhoun counties that served as a control area (JC).
[B] All concentrations below the LOD were substituted with LOD/$\sqrt{2}$.
[C] TEQ calculated based on WHO 2005 TEFs. All results are reported as parts per trillion (ppt) on a dry weight basis.

TABLE V. Summary of TEQ in House Dust Overall and for the Five Study Regions

	TEQ (ppt)[A]							
Study Region	Number	Mean	Standard Error	Median	75th Percentile	95th Percentile	Minimum	Maximum
Overall	764	36.7	4.3	16.2	29.7	126	1.4	1750
Floodplain	207	39.0	6.9	16.4	36.3	108	2.3	1750
Near floodplain	159	16.5	1.6	11.3	20.1	41.7	1.4	189
Elsewhere Midland/Saginaw	163	35.0	6.5	17.6	28.5	87.7	1.6	1400
Midland Plume	37	38.8	6.1	31.3	41.9	90.7	8.2	334
Jackson/Calhoun	198	38.8	6.1	13.8	32.2	177	2.1	1110

[A] TEQ calculated based on WHO 2005 TEFs.

contamination of house dust. The home was built in the early 1970s and is located in the floodplain of the Tittabawassee River. The property was flooded once by the Tittabawassee River, but the flood waters never entered the living area of the home (where dust samples were obtained), only the basement.

The occupants have lived in the home since the mid-1980s. They have no children and no pets, and they always take off their shoes when entering the home. They have never smoked in the home. Although no soil samples were obtained from this floodplain property, the TEQ of the house dust was dominated by furans (84.8% of TEQ from PCDFs) and appears to be consistent with the pattern of contamination in soil from the floodplain of the Tittabawassee River.[5]

Another possible source of house dust contamination from outside the home is secondary contamination from the occupation of an inhabitant. Occupational histories were obtained for all study participants in the baseline interviews and for all other home occupants as part of the follow-up interviews (Table III). No information was available concerning occupations of prior owners/occupants.

In two cases, an occupant had been employed in a setting that may have afforded opportunity for secondary contamination of the house dust with dioxin-like chemicals. In Case 1, an occupant had worked in a large metal machining operation for many years until the late 1990s, including occasional work on hydraulic machines. This individual retired more than 5 years prior to initial dust sampling, and no work clothes were available for analyses.

It is known that PCBs have been used in hydraulic oils.[14] TEQ in house dust (150 ppt) was dominated by PCBs (74.8% – Table II). In Case 6, an occupant had worked for a chemical company for approximately 30 years and had retired 3–6 years prior to dust sampling. Again, no work clothes were available for analyses. The TEQ for house dust (268 ppt) was roughly co-dominated by PCBs (51.1%) and PCDDs (41.5%). It is difficult to determine with certainty whether house dust was contaminated secondarily from the work place of these two subjects. In both cases, the pertinent employment had ended years previously, and work clothing was not available for testing.

Also, as shown in Table II, most contamination of house dust with PCDDs and/or PCBs was found in homes in which no one had potential exposure at work. Overall, it is uncertain, but seems less likely, that house dust contamination in these cases occurred via secondary contamination from employment.

Cigarette smoking or ashes from cigarettes were investigated as a possible source of contamination of house dust. There was a history of regular smoking inside the home in more than half of the dust outlier cases (Table III). However, the congener distributions in house dust of cases with smokers varied, with the TEQ in some homes dominated by PCBs and some dominated by PCDDs. In one case (Case 12), cigarette ash was sampled, and the total TEQ was low (7.81 ppt, Table IV). Overall, it appears unlikely that cigarettes are an important contributor to contamination of house dust with dioxins.

Wood-burning fireplaces were present in the homes of 8 of the 20 outlier cases, although in one case it had not been used since 1965 (Table III). Among the 8 cases with wood-burning fireplaces, the TEQ was dominated by PCBs in 6 cases (Cases 1, 2, 5, 6, 9, and 11), and by PCDDs in two cases (Cases 8 and 18) (Table II). Although PCBs can be produced by combustion of wood in home fireplaces, the contribution of PCBs to the overall TEQ of home fireplace emissions is small (approximately 0.5%), and so it is unlikely that fireplace emissions explain the high TEQ in house dust in these six cases.[15] Fireplaces cannot be ruled out as sources of contamination in the two cases in which the TEQ in dust was dominated by PCDDs, but it should also be noted that there were 7 other cases in which the dust contamination was dominated by PCDDs and that did not have fireplaces. Overall, it appears that wood-burning fireplaces are an unlikely source of contamination of house dust among the outlier cases in the present study.

There had been past structural fires in two cases (Cases 13 and 18). As noted above for Case 13, the house dust appears to have been contaminated with soil from the floodplain of the Tittabawassee River, and so it is unlikely that combustion products from a structural fire contributed to the contamination found in house dust. Case 18 is located in Jackson and Calhoun counties, and the original home was over a century old, with additions in the 1930s and 1960s. Sometime prior to the 1960s there was a fire in the coal bin in the basement just below the hallway where the dust samples were obtained. The hallway carpet was installed in the mid-1960s. The TEQ in house dust was 1110 ppt (93.6% PCDDs; 4.4% PCDFs; and 2.0% PCBs – Table II). A sample of charred wood from an original joist in the basement over the coal bin was analyzed, and the TEQ was 19.1 ppt (36.9% PCDDs; 31.9% PCDFs; 31.3% PCBs – Table IV). Overall, it appears unlikely that structural fires were the source of dioxin contamination of house dust in these two cases.

Repeat dust sampling was completed in 12 of 13 eligible cases. When the baseline samples were obtained from more than one location in the home, the repeat samples were obtained from the same locations but were analyzed separately. In 8 of 12 cases, dioxin levels in one or both repeat samples were elevated above the 95th percentile for house dust (TEQ = 126, Table V) and had roughly the same congener distributions as the baseline samples (supplemental Table V). The exceptions were Cases 6, 12, 13, and 15. No explanation for the decline in dioxin contamination of house dust for Cases 6 and 12 is apparent; the carpets had not been professionally cleaned in the interval between dates when the dust samples were obtained. The baseline dust for Case 13 had a TEQ = 411 ppt with 84.8% from PCDFs, while the follow-up dust had a TEQ = 25.3 ppt that was 76.1% PCDDs. It is notable that this was the only dust outlier home that had been officially remediated by Dow during the interval between baseline and follow-up dust sampling. Remediation in this case included commercial carpet cleaning of the areas that had been sampled for dust. It appears that the remediation reduced the contamination of the house dust in this case. Case 15 is similar in that the respondent reported that the carpets were routinely cleaned professionally on an annual basis, though not as part of a remediation effort by Dow. At least one or two professional cleanings had been performed after the first sample but before the second house dust samples were obtained. It appears that professional carpet cleaning is likely the reason for the decline in dioxin contamination of house dust in Case 15.

Seven cases had two separate dust samples analyzed at follow-up. In four cases, the TEQ in both samples were elevated and were within a factor of 2–3 of each other (Cases 1, 6, 12, and 15 – supplemental Table V), suggesting that the house dust contamination was generalized or relatively uniform within the house. In three cases, the TEQ of the two separate follow-up dust results were both elevated, but they differed by more than an order of magnitude (Cases 17, 18, and 19 – supplemental Table V), suggesting that the house dust contamination in these homes was more localized. In Case 17, the owner gave permission to sample a piece of carpet pad (from underneath the carpet with the more highly contaminated dust sample) for chemical analyses, and the results are shown in Table IV. The carpet pad and overlying carpet were at least 20 years old, but the exact ages were unknown. The high TEQ in the carpet pad (3950 ppt) with the congener profile dominated by PCBs suggested that the carpet pad was the source of the contamination of the dust. Case 18 is discussed above. In Case 19, a wood sample from an outdoor deck was sent for chemical analyses (Table IV); however, wood from the deck did not appear to be a source of contamination of the house dust in this case.

For Case 20, the home is located in the floodplain of the Tittabawassee River (within 200 meters of the river) and was newly built and decorated in the late 1990s with all new interior furnishings (i.e., carpets, curtains, furniture, paint, etc.). It has been occupied by only one owner. The TEQ in house dust was 1750 ppt (98.7% PCDDs; 1.2% PCDFs; 0.1% PCBs—Table II), and the congener distribution was not consistent with contamination found in soil in the floodplain, but no soil samples were obtained for analyses from this property.

There was a large leather-covered sectional sofa in the living room that was purchased new from a regional chain furniture retailer at the time the home was first occupied. The owner recalled that the leather was imported but could not recall from where. Pentachlorophenol (known to be contaminated with PCDDs) has been used in leather processing.[16] A sample of leather was sent for chemical analyses, and the TEQ was 61.3 ppt (66.4% PCDDs, 23.5% PCDFs, and 10.1% PCBs—Table IV). It appears unlikely that the leather from the sofa was the source of contamination of house dust in this case.

DISCUSSION

Outlier contamination of house dust with dioxins was found even in areas without known environmental contamination. In 2 of 20 outlier cases, contaminated soil from the property was the likely source of contamination of house dust (Cases 5 and 13); soil from around the home was most likely not the source of contamination in the other cases. In one case, there was an identifiable source of contamination inside the home (Case 17—carpet pad). In two homes (Cases 1 and 6), it was possible, though less likely, that house dust was secondarily contaminated from occupational exposures of occupants. Fireplaces were unlikely sources of contamination of house dust in the present study.

One outlier case (Case 10) was located in the Midland Plume area, the region in the parent study that was likely contaminated via historical aerosol deposition from incineration activities at Dow (and was the region that had the highest median level of dust contamination). House dust and soil contamination among houses in the plume area was dominated by PCDDs (~70% and ~80% of the TEQ, respectively), but the contamination in house dust in Case 10 was dominated by PCBs (86% of TEQ), and so it appears unlikely that the dust contamination in this case was related to aerosol deposition from incineration activities at Dow. None of the other dust outlier cases was located near or downwind from a known point source of dioxin aerosol emissions. In most cases the origin of contamination of house dust could not be determined but appears likely to be from sources inside the homes.

Before use was stopped, there were various consumer products that were made with PCBs, including fluorescent light fixtures, electrical devices or appliances containing PCB capacitors, paints, caulks and plasticizers for various products (e.g., ceiling tiles).[14,17] Homeowners are unlikely to know whether such PCB-contaminated products are present in homes, and so, such products could be sources of emissions from within homes, and would be "uncharacterized." We documented one example (Case 17, involving a carpet pad), but we strongly suspect that others exist.

In contrast, PCDDs were never produced intentionally as commercial products; they are produced unintentionally in various chemical processes involving chlorine and also certain incineration and combustion processes.[16] However, PCDDs are contaminants in various commercial products, such as certain pesticides. It is possible that pesticides or other products contaminated with PCDDs may be incorporated in materials used in homes (e.g., leather), although we were unable to confirm any instances. Again, homeowners are unlikely to be aware of such contamination, and so, emissions from such sources in homes would be uncharacterized.

Somewhat surprisingly, the largest number of dust outliers was from Jackson/Calhoun counties ($n = 9$ out of 20), the control area for the study. The congeners contributing most to the TEQ among outliers varied, with some cases dominated (i.e., >50% of the TEQ) by PCDDs ($n = 9$), PCDFs ($n = 1$) and PCBs ($n = 9$), respectively, which indicates that the sources of house dust contamination are variable. It is interesting to note that most (6 out of 9) outlier cases dominated by PCDDs were from study regions in Midland and Saginaw counties, whereas most outlier cases dominated by PCBs (6 out of 9) were from Jackson and Calhoun counties (Table II).

The soil of 21 homes in the UMDES had a maximum soil TEQ $\geq$ 1000 ppt, including 19 homes in the floodplain, but none of these homes had outlier dioxin levels in house dust. Note that all baseline house dust samples were obtained prior to remediation of frequently flooded homes in the floodplain of the Tittabawassee River. All house dust outliers for which soil measurements were available had house perimeter and maximum soil TEQs less than 75 ppt (Table II). Factors that may have caused outlier house dust contamination in some but not most cases with contaminated soil are unclear (e.g., presence of indoor/outdoor pets, taking off shoes inside the home, presence of children, intrinsic properties of different soils, other behavioral factors, etc.).

It was possible to identify a likely source of contamination of house dust from inside the home in one case (i.e., Case 17—carpet pad). This house was old, and the carpet and pad were of unknown age, but over 20 years old, which confirms that in the past some home furnishings and/or construction materials were made or contaminated with dioxin-like chemicals, particularly PCBs.[14,17] However, even recently constructed homes with all new furnishings apparently can have high dioxin contamination in house dust (e.g., see Case 20), which suggests that some contemporary interior home furnishings are still manufactured in ways that incorporate dioxin-like chemicals and that these products can release dioxins that are detectable at high levels in house dust.

The dioxin contamination in follow-up house dust samples was reduced from baseline in four cases. In two cases (Cases 6 and 12), there was no clear explanation for the reduction in dioxin contamination of house dust. The reduction appeared to be the result of professional carpet cleaning in two other cases (Cases 13 and 15). In Case 4, the carpets were reported to have been cleaned professionally between the house dust sampling campaigns, but the level of dioxin contamination in house dust was not reduced substantially in this case. Further research is needed to determine whether professional carpet cleaning is effective in reducing dioxin contamination of house dust. A number of factors appeared not to be important contributors to house dust contamination in the present study, including cigarette ash/smoking in the home, a history of a structural fire, presence of a wood deck and leather from a sofa.

Most previous studies of dioxin contamination of house dust obtained via vacuum sampling have been limited in terms of the number of cases studied and spectrum of chemicals analyzed. Berry et al.[1] measured PCDDs ($n = 7$) and PCDFs ($n = 10$) in only two samples of house dust collected in vacuum cleaner bags from homes in Canada; there were no analyses for PCBs. The concentrations of dioxins in house dust, based on International TEQs (I-TEQs)[18] were 8.3 ppt and 12 ppt per gram of dust. There was no description of the location of the home(s) from which dust samples were obtained in relation to potential environmental contamination, and no measurements of soil adjacent to the homes.

Wittsiepe et al.[4] collected house dust from conventional vacuum cleaner bags from 22 homes in Germany: 10 homes believed to be uncontaminated; 10 homes believed to be contaminated because they were located near a former metal reclamation plant; and 2 old farm houses that had been treated with pentachlorophenol (PCP) some years earlier. Dust samples were analyzed for PCDDs and PCDFs, not PCBs. The mean I-TEQs in dust were 101 ppt and 265 ppt for the 10 uncontaminated and 10 homes suspected of contamination, respectively. The I-TEQs in dust from the two PCP-treated farm houses were 1390 ppt and 11,800 ppt. Although no data were shown, the authors compared the TEQ and congener patterns of house dust to soil and concluded that, on average, house dust was more contaminated than soil, and that the congener patterns tended to differ. They also concluded that outdoor dust or soil did not appear to be an important contributor to contamination of indoor dust and that, as in the present study, indoor sources were probably more important contributors to contamination of house dust. It was conjectured that indoor sources of contamination of house dust might include textiles (clothes, carpets, upholstery, etc.), leather goods, and pentachlorophenol-treated furniture.

Saito et al.[3] collected house dust samples via vacuuming from five homes in each of two cities in Japan. The precise method of dust collection is not described. The mean TEQ for the five homes from each city, based on the 29 PCDDs, PCDFs and PCBs with TEFs (using WHO TEFs from 1998),[7] were 15.6 ppt and 16.0 ppt, respectively. They concluded that the dioxin levels in house dust were similar to the levels of contamination in soil in Japan.

O'Connor and Sabrsula[2] reported results of measurements of PCDDs and PCDFs (no PCBs were measured) in 14 house dust samples collected from 12 homes in a single community in rural Mississippi. This community was selected because it was not believed to be unduly polluted by airborne dioxins. A total of 34 dust samples were obtained from 18 homes, but 20 samples were discarded because of suspected contamination with sand, carpet powders, or household sprays. Of the remaining 14 dust samples, 6 consisted of carpet dust obtained with conventional home vacuum cleaners, and 8 dust samples reflected general household dust (e.g., dust from undisturbed areas) obtained using special hand-held vacuum cleaners fitted with HEPA filters.

Two homes had paired samples: one conventional vacuum cleaner dust sample and one dust sample obtained using the hand-held vacuum. The mean TEQ (based on 1998 WHO TEFs)[7] of the 14 dust samples was 20.3 ppt (standard deviation 18.4 ppt, range 1.30–53.7 ppt). Results from the two collection methods appeared to be comparable. The authors noted that dioxin levels in house dust did not appear to be related to the presence of a fireplace in the home, which is in agreement with the present study.

Rudel et al.[19] collected vacuum samples of house dust from 119 homes of women with breast cancer or matched controls living on Cape Cod, Massachusetts. The dust samples were analyzed for 86 different target compounds, including 3 PCB congeners (PCB 52, PCB 105, and PCB 153) but no PCDDs or PCDFs. Only PCB 105 has been assigned a TEF by the

WHO, and so this is the only one of the three congeners to be reported here.[8] Rudel et al. found PCB 105 above their limit of detection (LOD) (~0.2 micrograms per gram of house dust or 200,000 ppt) in 9 (7.6%) of the 119 homes. The median LOD for PCB 105 in the present study was 2.1 ppt. PCB 105 was detectable above the LOD in 763 out of 764 house dust measurements; the 95th percentile for PCB 105 in house dust was 33,800 ppt, and the concentration of PCB 105 in house dust was greater than 200,000 ppt in 9 (1.2%) of 764 cases. Eight of these 9 cases are among the outliers described in this report (Cases 1, 2, 3, 4, 9, 10, 11, and 17 – supplemental Table II).

CONCLUSIONS

Outlier dioxin contamination of house dust, i.e., dust with a TEQ more than 2.5 standard deviations above the mean of the log-transformed data, was found in 20 of 764 homes. In 2 of 20 outlier cases, house dust contamination appeared to be related to contamination in adjacent soil; in most cases, house dust contamination in homes with outlier values was not related to contamination in adjacent soils. Extreme contamination of adjacent soil (i.e., TEQ $\geq$ 1000 ppt) usually did not result in outlier levels of contamination of house dust. In one case, the source of house dust contamination could be reasonably attributed to a source inside the home (a carpet pad contaminated with PCBs). In two cases, there was possible (though unlikely) secondary contamination of house dust from prior occupational exposures of occupants. The contamination in dust outlier cases appeared to be unrelated to known environmental emission sources of dioxin contaminated aerosols. In most outlier cases, the origins of house dust contamination were not identified but appear to be related to uncharacterized sources within homes. Based on a limited number of cases, professional cleaning of carpets may be effective in reducing dioxin contamination in house dust. More research is needed to identify indoor sources of dioxin contamination within homes.

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