

A FOLLOW-UP INVESTIGATION OF HOMES WITH 'HIGH' CONCENTRATIONS OF PCDDs, PCDFs AND DIOXIN-LIKE PCBs IN HOUSE DUST

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Abstract

As part of the University of Michigan Dioxin Exposure Study (UMDES), the 29 congeners of polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and dioxin-like polychlorinated biphenyls (PCBs) that have consensus toxic equivalency factors (TEFs) were measured in house dust of 764 subjects who were a representative sample of the general population in five Michigan counties. Thirteen subjects with the highest toxic equivalency (TEQ) in house dust, and who were otherwise eligible, were re-interviewed as part of an exploratory investigation to determine why these results were elevated. Likely sources of contamination of house dust were identified in only a minority of cases.

Introduction

Few studies have measured PCDDs, PCDFs and/or dioxin-like PCBs in house dust.^{1, 2, 3, 4} All of these previous studies have been small (fewer than 15 house dust samples in each study). No study has attempted to obtain and analyze house dust samples from a large, representative sample of homes most of which are not suspected of contamination. The University of Michigan Dioxin Exposure Study (UMDES) was designed to determine whether PCDDs, PCDFs, and dioxin-like PCBs (hereinafter 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., diet, occupation, age, body mass index, 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, USA, have contaminated areas of the City of Midland and sediments in the Tittabawassee River flood plain. The study measured the levels of the World Health Organization 29 dioxin congeners with consensus TEFs in serum, soil and house dust from a random sample of the population in the study regions. Not surprisingly, the distribution of TEQs in house dust is skewed, with a small number of high or 'outlier' values. This follow-up study describes these results and explores what may explain high TEQ values in house dust.

Materials and Methods

To be eligible for this follow-up study, a person must have been a participant in the UMDES, and must have completed an interview and permitted collection of a sample of house dust for chemical analyses. Full details on UMDES study protocols are described elsewhere (<http://www.sph.umich.edu/dioxin/protocol.html>). The total TEQ of the house dust sample must have been an 'outlier', as defined below. Subjects were given the option to receive the results and 67% of subjects in the original study elected to receive results of chemical analyses of their house dust. Only subjects who received results of chemical analyses of house dust were eligible to participate in this follow-up study (e.g., if a subject had an 'outlier' dust result, but he chose to not receive his dust results, then he was not eligible to be recruited to participate in the follow-up study). There were a total of 764 subjects with house dust samples: 205 from the floodplain of the Tittabawassee River (FP), 161 from the near-floodplain of the Tittabawassee River (NFP), 32 from the Midland plume area downwind from the historic incineration activities of the Dow plant (PL), 168 from elsewhere in Midland and Saginaw counties (MS), and 198 from Jackson and Calhoun counties that served as a control area (JC). A dust outlier was defined as one with a total TEQ that was more than 2.5 standard deviations (sd) above the mean of the log-transformed data [Note: this investigation was initiated in mid-2005, and the determination of 'outlier' status was based on

calculation of the TEQs using the 1997 TEFs; all TEQ results shown have been re-calculated using the 2005 TEFs⁵]. There were 20 house dust results that met this statistical criterion, and 13 of the subjects had elected to receive results and were therefore eligible to participate in the follow-up study. Interviews were completed with all 13 eligible subjects. The follow-up interview included open-ended questions covering the following topic areas: house characteristics (e.g., age of house, age of carpets/rugs, fireplace use, trash burning, flooding, remediation, professional cleaning of floors/rugs/carpets, use of pesticides inside or outside the house, history of renovations or structural fires, nearby industries); characteristics/habits of occupants of the house (e.g., ages of occupants, pets, 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). Given space limitations, only the most pertinent interview results are summarized below. In four cases non-dust materials were sent for chemical analyses, as described below. This study was reviewed and approved by the University of Michigan Health IRB.

Results and Discussion

The overall mean and median TEQ values for the 764 house dust measurements were 36.7 parts per trillion (ppt) and 16.2 ppt, respectively. Congener-specific results of chemical analyses of house dust for the 13 outlier cases are listed in Table 1. Individual case summaries follow:

Case 1: Location: FP. Home was built in early 1960's. Remote smoking by one occupant (>25 years ago). Carpets purchased 10-15 years prior to dust sampling in 2005. One occupant had worked in a foundry for many years until late 1990's. TEQ in house dust: 150 ppt (2.5% PCDDs; 22.7% PCDFs; 74.9% PCBs). The TEQ in soil from the property: 31.8 ppt (57.3% PCDDs; 37.9% PCDFs; 4.8% PCBs).

Case 2: Location: JC. Home was built in the 1920's. Occupants do not smoke; relatives visit and smoke often. New carpets installed 10-12 years prior to dust sampling. TEQ in house dust: 177 ppt (26.1% PCDDs; 16.7% PCDFs; 57.2% PCBs). TEQ in soil from the property: 13.9 ppt (42.8% PCDDs; 19.7% PCDFs; 37.5% PCBs).

Case 3: Location: JC. Home was built in the 1920's. Occupant does not smoke, but significant other smokes. New carpets installed in the early 1990's. TEQ in house dust: 199 ppt (3.4% PCDDs; 5.2% PCDFs; 91.4% PCBs). TEQ in soil from the property: 7.5 ppt (49.5% PCDDs; 26.6% PCDFs; 23.9% PCBs).

Case 4: Location: JC. Home was built in the early 1960's. No cigarette smoking. New carpets were installed in the 1980's. The occupants have maintained a flower garden just outside the front door of the house; every year they have added multiple bags of top soil purchased from local 'box' store retailers. TEQ in house dust: 224 ppt (8.6% PCDDs; 5.0% PCDFs; 86.4% PCBs). TEQ in soil from the flower garden: 72.8 ppt (2.1% PCDDs; 3.2% PCDFs; 94.7% PCBs). TEQ in soil from house perimeter: 7.9 ppt (16.6% PCDDs; 13.3% PCDFs; 70.1% PCBs).

Case 5: Location: MS. Home was built in the mid-1960's. No smoking. All carpets (6 years old) and rugs (3-4 years old) purchased new. One occupant had worked for a chemical company for about 30 years, and had retired 3-6 years prior to dust sampling. TEQ in house dust: 268 ppt (41.4% PCDDs; 7.4% PCDFs; 51.1% PCBs). TEQ in soil from the property: 8.7 ppt (79.5% PCDDs; 18.2% PCDFs; 2.4% PCBs).

Case 6: Location: MS. Home is more than 90 years old. One occupant smokes; some visitors smoke. New carpets installed about 20-25 years prior to dust sampling. TEQ in house dust: 334 ppt (4.2% PCDDs; 9.8% PCDFs; 86.1% PCBs). TEQ in soil from the property: 60.6 ppt (70.4% PCDDs; 26.6% PCDFs; 2.9% PCBs).

Case 7: Location MS. House was built in the early 1960's. New carpets and rugs were installed in the 1990's. The occupants smoke. TEQ in house dust: 409 ppt (88.5% PCDDs; 6.6% PCDFs; 4.9% PCBs). TEQ in soil from the property: 5.2 ppt (40.3% PCDDs; 45.5% PCDFs; 14.2% PCBs). TEQ in cigarette ash: 7.8 ppt (79.5% PCDDs; 14.6% PCDFs; 5.9% PCBs).

Case 8: Location FP. Home was built in the early 1970's. No smoking in the house. TEQ in house dust: 408 ppt (11.7% PCDDs; 85.6% PCDFs; 2.7% PCBs). No soil samples were available, but the congener pattern of the house dust was similar to the contamination in the flood plain.

Case 9: Location JC. Home was built in the 1940's. No smoking in the house. New carpets installed in 2000. TEQ in house dust: 544 ppt (93.2% PCDDs; 6.6% PCDFs; 0.2% PCBs). TEQ in soil from the property: 5.3 ppt (30.4% PCDDs; 30.4% PCDFs; 39.3% PCBs).

Case 10: Location MS. Home was built during the first decade of the 20th century. Many people have smoked in the house. Carpet age is unknown, but over 20 years. TEQ in house dust: 824 ppt (49.3% PCDDs; 15.9%

PCDFs; 34.8% PCBs). TEQ in soil from the property: 19.1 ppt (34.8% PCDDs; 40.9% PCDFs; 24.3% PCBs). TEQ in sample of carpet and pad: 3950 ppt (27.3% PCDDs; 6.6% PCDFs; 66.2% PCBs).

Case 11: Location JC. Original home was built in the late 19th century, with additions in the 1930's and 1960's. Sometime prior to the 1960's there was a fire in the coal bin in the basement, just underneath the hallway from which dust samples were obtained. The hallway carpet was installed in the mid-1960's. Smokers occupy the house. TEQ in house dust: 1110 ppt (93.6% PCDDs; 4.4% PCDFs; 2.0% PCBs). TEQ in soil from the property: 19.1 ppt (70.9% PCDDs; 21.3% PCDFs; 7.8% PCBs). A sample of charred wood from an original joist from the basement was also analyzed for dioxins: TEQ = 19.1 ppt (37.9% PCDDs; 31.9% PCDFs; 31.3% PCBs).

Case 12: Location MS. Age of home is unclear, but has been occupied by current residents since the early 1960's. No cigarette smoking. New carpet was installed 10-15 years prior to sampling. A wood deck was added about 10 years ago (unclear if pressure treated wood). TEQ in house dust: 1400 ppt (93.4% PCDDs; 6.4% PCDFs; 0.2% PCBs). TEQ in soil from the property: 15.4 ppt (27.3% PCDDs; 60.3% PCDFs; 12.4% PCBs). TEQ from wood sample from deck: 16.0 ppt (75.7% PCDDs; 16.6% PCDFs; 7.8% PCBs).

Case 13: Location FP. Home was built in the late 1990's, with all new interior furnishings (i.e., carpets, curtains, furniture, paint, etc.). Occupied by nonsmoker; some visitors smoke. TEQ in house dust: 1750 ppt (98.7% PCDDs; 1.2% PCDFs; 0.1% PCBs). No soil samples available.

Outliers were located in all regions except the plume area; the largest number was from Jackson/Calhoun (n=5 out of 13). The congeners contributing most to the TEQ among outliers varied, with some cases dominated by PCDDs (n=6), PCDFs (n=1) and PCBs (n=6), respectively. Soil contamination appeared to be a dominant factor for contamination of house dust in a minority of cases (see cases 4 and 8). Just as intriguing was that many homes with 'high' soil contamination did not have correspondingly 'high' dioxin levels in house dust (not shown). Occupant factors that may have contributed to these contrasting relationships between soil and house dust contamination are unclear (e.g., outdoor pets, taking off shoes inside the home, presence of children, etc.). Even recently constructed homes can have high dioxin contamination in house dust (e.g., see case 13). In one case it was possible to identify a likely source of contamination of house dust from a source inside the home (e.g., case 10 – carpet and/or pad). Cigarette ash has a low TEQ (see case 7) and appears to not be an important contributor to the dioxin contamination of house dust. In two cases (cases 1 and 5) residents had occupations that may have afforded opportunity for exposures to dioxin-like chemicals and secondary contamination of house dust, however, the congener profiles in house dust in these 2 cases did not appear to differ conspicuously from other cases in which there was no suspicious occupational history. A remote structural fire did not appear to be contributing factor to current elevation of dioxins in house dust (see case 11). In most cases the dominant source(s) of house dust contamination in homes with 'outlier' values were not related to adjacent soil; in many or most such cases the source(s) of contamination are unknown, but may be related to uncharacterized sources within homes.

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Table 1: Baseline Concentrations of PCDDs, PCDFs, and dioxin-like PCBs in House Dust (picograms/gram of house dust dry weight)

Case Number/Location	1 – FP	2 – JC	3 – JC	4 – JC	5 - MS	6 – MS	7 – MS	8 – FP	9 – JC	10 – MS	11 – JC	12 – MS	13 - FP
2,3,7,8-TCDD	0.22**	0.12**	0.21**	0.28**	0.78**	4.06	5.82	2.83	0.27**	1.4**	2.37	0.48	0.74**
1,2,3,7,8-PeCDD	0.44**	0.45**	1.03	1.07**	9.27	3.68	13.2	5.8**	0.76**	6.16	7.32	3.36	4.41
1,2,3,4,7,8-HxCDD	0.71	6.46	1.43	4.05	19.2	4.02	37.4	16.3**	23.3	20.1	46.9	105	3.36**
1,2,3,6,7,8-HxCDD	4.97	129	12	35.4	107	12.2	397	59.5	521	533	1460	2120	13200
1,2,3,7,8,9-HxCDD	2.41	20.3	4.24	8.91	47.1	2.36	157	16.8**	90.1	133	230	438	2230
1,2,3,4,6,7,8-HpCDD	194	2650	319	1080	6950	340	23800	2420	38300	27700	77700	97100	17000
OCDD	943	11400	1960	7610	47700	3020	152000	19200	198000	176000	274000	223000	21500
2,3,7,8-TCDF	4.87	4.92	4.13	4.43	8.8	49.9	26.8	991	0.75	19.3	5.6	4.45	2.94
1,2,3,7,8-PeCDF	5.03	3.02	2.23	2.65	5.41	19.4	16	579	1.78	16.4	3.72	3.5	2.53
2,3,4,7,8-PeCDF	57.4	32.7	22.6	10.9	12.6	63.7	27.4	512	3.36	105	14.4	16.2	3.17
1,2,3,4,7,8-HxCDF	90.6	85.7	12.9	30	18.5	34.9	28.6	401	20.7	512	41.5	206	13
1,2,3,6,7,8-HxCDF	39.5	39.7	5.79	20.5	36.3	15.5	33.9	105	52.9	221	31.9	81.5	27.2
1,2,3,7,8,9-HxCDF	9.57	15	2.37	0.59**	4.59	6.56	7.44	78.7	0.77**	57.1	8.82	43.4	0.78**
2,3,4,6,7,8-HxCDF	17.3	23.3	4.56	8.45	29.2	12.9	32.7	73.1	45	83.7	57.7	134	31.8
1,2,3,4,6,7,8-HpCDF	39.3	219	49.7	131	541	88.1	472	1220	2040	800	2560	3230	1240
1,2,3,4,7,8,9-HpCDF	10	40.2	4.75	8.7	26.9	11.6	26.3	64.3	53.3	131	205	333	13
OCDF	33.8	689	108	252	1920	164	713	1840	6520	1940	7930	8950	1160
PCB 81	87.5	84.5	4970	173	253	5480	168	87.4	3.33	178	43.9	30	8.07
PCB 77	2510	2380	85800	5520	9040	91500	3370	1710	77.8	5400	1250	525	190
PCB 126	820	600	1520	1750	1340	2580	140	91.9	7.7	2010	174	22.1	11.1
PCB 169	21.9	16.9	3.92	47.2	31.6	6.16	9.78	1.26**	1.48	87.7	7.46	1.29	1.92
PCB 105	245000	381000	244000	115000	12400	234000	45600	12400	515	661000	43800	3930	1630
PCB 114	14800	36000	19500	6710	496	24800	2760	828	27.2	51900	2050	271	95.8
PCB 118	532000	608000	362000	357000	22800	315000	110000	27700	1200	1170000	86500	9540	4230
PCB 123	6480	13600	13800	4450	387	19800	1810	1160	30.7	26700	1400	348	58.5
PCB 156	121000	189000	13600	48000	3600	14800	10300	4470	167	471000	15600	1140	1170
PCB 157	27100	42300	2910	11500	957	4110	2240	927	29.8	124000	3240	266	121
PCB 167	38900	62700	4100	16800	1200	4670	3580	1640	73.5	193000	4850	463	432
PCB 189	4270	8060	1040	1880	136	579	445	850	25.7	45500	698	68.7	245
Total TEQ (ppt)	150	176	199	224	268	334	408	411	544	824	1110	1400	1750

All concentrations below the Limit of Detection (LOD) were substituted with LOD/ $\sqrt{2}$ (denoted by **). 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)