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# Determinants of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans in house dust samples from four areas of the United States

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# Abstract

Determinants of levels of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/F) in dust in U.S. homes are not well characterized. We conducted a pilot study to evaluate the relationship between concentrations of PCDD/F in house dust and residential proximity to known sources, including industrial facilities and traffic. Samples from vacuum bag dust from homes of 40 residents of Detroit, Los Angeles, Seattle, or Iowa who participated in a population-based case-control study of non-Hodgkin lymphoma conducted in 1998-2000 were analyzed using high resolution gas chromatography/high resolution mass spectrometry for 7 PCDD and 10 PCDF congeners considered toxic by the U.S. Environmental Protection Agency (EPA). Locations of 10 types of PCDD/F-emitting facilities were obtained from the EPA; however only 4 types were located near study homes (non-hazardous waste cement kilns, coal-fired power plants, sewage sludge incinerators, and medical waste incinerators). Relationships between concentrations of each PCDD/F and proximity to industrial facilities, freight routes, and major roads were evaluated using separate multivariate regression models for each congener. The median (inter-quartile range [IQR]) toxic equivalence (TEQ) concentration of these congeners in the house dust was 20.3 pg/g (IOR=14.3, 32.7). Homes within 3 or 5 km of a cement kiln had 2 to 9-fold higher concentrations of 5 PCDD and 5 PCDF (p<0.1 in each model). Proximity to freight routes and major roads was associated with elevated concentrations of 1 PCDD and 8 PCDF. Higher concentrations of certain PCDD/F in homes near cement kilns, freight routes, and major roads suggest these outdoor sources are contributing to indoor environmental exposures. Further study of the contribution of these sources and other facility types to total PCDD/F exposure in a larger number of homes is warranted.

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# Keywords

dioxins; furans; geographic information systems; dust; air pollution; environmental exposure; non-Hodgkin lymphoma (NHL)

# 1. Introduction

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/F) are persistent organic pollutants with similar structures and mechanisms of action. PCDD/F enter the environment mainly through the air as products of combustion, such as waste incineration, metal smelting, cement production, and vehicle exhaust (U.S.EPA, 2006). PCDD/F have been associated with cancer and other adverse health outcomes in populations exposed occupationally or via accidental release (IARC, 1997; Baan et al., 2009; Pesatori et al., 2009; NTP, 2011; Warner et al., 2011). However, the relationship between exposure to environmental levels of PCDD/F and risk of adverse health effects in the general population is unclear (Boffetta et al., 2011). There is epidemiologic evidence to suggest an increased risk of non-Hodgkin lymphoma (NHL) among people living hear facilities that emit dioxins to the air (Floret et al., 2003; Viel et al., 2008b).

The National Cancer Institute Surveillance, Epidemiology and End Results Non-Hodgkin Lymphoma (NCI-SEER NHL) Study is a population-based case-control study conducted in four areas of the United States to evaluate environmental and other risk factors for NHL. We previously examined the association between NHL risk and residential proximity to PCDD/ F-emitting facilities in the NCI-SEER NHL study using an average emission index (AEI) that weights PCDD/F emissions from one or more facilities by the inverse of the squared distances to the home (Pronk et al., submitted). We found no association between the AEI and NHL risk, but we did observe an increased risk of NHL (OR=3.8, 95% CI 1.1–14.0) among people living within 3 km of cement kilns. To better understand these epidemiologic findings, we conducted a pilot study to evaluate the AEI and other exposure metrics used in this study in relation to measured levels of PCDD/F in carpet dust.

Carpet dust acts as a reservoir for many environmental pollutants and may integrate exposure from multiple sources (Butte and Heinzow, 2002). Higher concentrations of PCDD/F were found in dust sampled from homes located downwind of a former dioxinemitting incinerator and pesticide manufacturer compared to homes in a reference county approximately 100 miles away (UMDES 2008). Some other studies have investigated PCDD/F in house dust, but have had relatively small sample sizes (<25 homes) (Berry et al., 1993; Wittsiepe et al., 1997; Saito et al., 2003; O'Connor and Sabrsula, 2005) or have targeted homes near a single industrial facility with known or suspected PCDD/F releases to the surrounding environment with no comparison group (Dahlgren et al., 2007; Gonzalez et al. 2011; Feng et al., 2011; Hensley et al., 2007). Our pilot study compares residential proximity to multiple industrial facility- and traffic-related PCDD/F sources to measured concentrations of PCDD/F in dust from 40 homes in four areas of the United States.

# 2. Methods

# 2.1. Study Population

We selected 40 participants from the NCI-SEER NHL Study. As previously described in detail (Colt et al., 2005; De Roos et al., 2010), 1,321 cases and 1,057 controls from Iowa, Los Angeles County, Detroit, and Seattle were interviewed from 1998 to 2000 in their homes using a computer assisted personal interview that included questions about demographics, residential and occupational histories, and other factors. A total of 1,106

participants who had used their vacuum cleaner within the prior year and had owned at least half of their carpets or rugs for at least 5 years provided the dust from their vacuum bags at the time of interview. The dust was previously analyzed for several classes of chemicals, including polychlorinated biphenyls (PCBs), pesticides, and polycyclic aromatic hydrocarbons (Colt et al., 2005). Of these 1,106 participants, 98 had plasma samples that were previously analyzed for PCDD/F and evaluated for their association with risk of NHL (De Roos et al., 2005). For this pilot study, we considered eligible the 85 participants (47 cases and 38 controls) who had PCDD/F plasma measurements and sufficient vacuum dust available (4 g) for chemical analysis of PCDD/F. This pilot study focuses only on the dust samples; studies comparing the AEI and dust PCDD/F measurements to plasma PCDD/F measurements are planned. The residence of eligible participants at the time of interview was classified into groups based on the AEI (greater than zero or equal to zero [described in more detail below]), population density of the census block (greater than or less than the median for the 85 eligible participants [median=3130 people per square mile]), and proximity to a major road (within 200 m, 201-400 m, greater than 400 m). Approximately equal numbers were sampled from each group such that all combinations of these variables were represented in our pilot study population (N=40). The median duration of residence in the interview home was 24 yrs (range: 4-50 yrs).

#### 2.2. Residence Locations

Current residential addresses provided by participants were geocoded using a modified Microsoft Visual Basic version 6.0 program (TeleAtlas, Lebanon, NH) to match input addresses to the TeleAtlas MatchMaker SDK Professional version 4.3 spatial database of roads. Additionally, interviewers took a global positioning system (GPS) reading outside the participant's current home using a Garmin GPS12 Personal Navigator (Garmin International, Inc., Olathe, KS). Discrepancies between GPS and geocoded coordinates 200 m were visually checked using mapping software (ArcMap, Esri, Redlands CA) and web-based aerial photographs (http://maps.google.com/). Of the 40 residential locations, 38 (95%) were quality-checked GPS coordinates, and 2 (5%) were geocoded coordinates.

# 2.3. Proximity to Industrial Facilities

Using Geographic Information Systems (GIS) analysis, we calculated the proximity of residences to PCDD/F emitting facilities. Addresses and coordinates of the industrial facilities were obtained from an Environmental Protection Agency (EPA) database (U.S.EPA, 2001) and verified by comparison to web-based aerial photographs and supplementary information. Facilities included secondary copper smelters, municipal solid waste incinerators, medical waste incinerators, sewage sludge incinerators, hazardous waste incinerators, cement kilns burning non-hazardous waste, cement kilns burning hazardous waste, iron ore sintering plants, coal-fired electric generating plants, and industrial boilers. Together these facility types comprised over 85% of estimated total dioxin air emissions in the United States over the past 30 years (U.S.EPA, 2006). We evaluated proximity of residences to PCDD/F-emitting facilities at distances up to 3 km and 5 km, based on reports of elevated levels of PCDD/F in soil up to approximately 3 km away from municipal solid waste incinerators (Domingo et al., 2000; Floret et al., 2003; Lorber et al., 1998; Viel et al., 2008b). We extended the distance to 5 km to account for potential longer distance transport from some facility types (personal communication, David Cleverly, U.S.EPA).

# 2.4. Average Annual Dioxin Emission Index (AEI)

We computed a dioxin emission index for the interview home (i.e., the residence in which the dust sample was collected) using the EPA database, as described in detail by Pronk et al. (submitted). Briefly, an annual estimated emission (described below) from each facility within 5 km (or 3 km) of a residence was divided by the square of the distance between the

facility and the residence and summed over all facilities within 5 or 3 km of the residence We averaged the annual dioxin emission index over the duration of residence in the interview home, up to 15 years (i.e., going back as far as 1983). We did not go back further in time due to limited information on emissions.

The annual emission was expressed in terms of a toxic equivalence (TEQ), a summed metric that weights PCDD/F congeners relative to the potency of the most potent congener, 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD), using WHO 1998 toxic equivalence factors (TEFs) (U.S.EPA, 2010; Van den Berg et al., 2006). Annual emissions data (ng TEQ/yr) for 1995 were available for all facility types in our study. We estimated emissions in other years based on information from an EPA national survey of dioxin-emitting facilities in 1987, 1995, and 2000 (U.S.EPA, 2006). We estimated the rate of change in emissions (TEQ ng/ year) between 1987 and 1995 for each facility type as the difference between the average emission level in 1987 and 1995 divided by 8 years. We used the same approach to estimate the rate of change in emissions between 1995 and 2000. We applied the relevant rate of change to a facility's 1995 emission level to estimate facility-specific emission levels between 1987 and 1996 and 2000. We assumed constant emission levels prior to 1987 when air pollution controls were uncommon.

# 2.5. Proximity to Traffic Sources of Dioxins

Using GIS analysis, we developed two traffic-related determinants: proximity to major roads and proximity to freight routes. Major roads had arterial classifications of inter-state, intermetropolitan area, or intra-state/intra-metropolitan area/inter-metropolitan area (TeleAtlas Dynamap Transportation version 5.2, 2003). Freight routes included any roads permitting trucks (Freight Analysis Framework 2.2 Network Machine Readable Data Files, Federal Highway Administration Office of Freight Management and Operations, Washington DC, 2009). Therefore, the two traffic-related variables were not mutually exclusive. Proximity was characterized by the distance from the residence to the nearest major road or freight route. Participants living more than 1000 m from a major road or freight route were assigned a distance of 1000 m.

# 2.6. Demographic and Residential Characteristics

We considered the following demographic, residential, and other characteristics as potential determinants of PCDD/F concentrations in house dust: study center; case-control status; sex; year residence built; smoking status; and occupation in mills, refineries, the refuse industry, or farming (all of these occupations have potentially high exposure to PCDD/F). Information on smoking, a potential indoor source of PCDD/F (Lofroth and Zebuhr, 1992; Wilson et al., 2008), was not ascertained in 45% of the pilot study population because of the split-sample design in the parent study. We also considered population density a risk factor for potential exposure (Cleverly et al., 2007) and calculated density (people per square mile, ppsm) based on Year 2000 population and area of the U.S. census block of the residences (U.S.Census Bureau, 2000). We divided the reported density (ppsm) by 1000 to facilitate interpretation of regression coefficients in our regression models.

# 2.7. Laboratory Analysis of House Dust

Vacuum bags were shipped to Southwest Research Institute (San Antonio, TX), sieved (<150  $\mu$ m), and frozen at -20 °C until analysis (maximum storage time=10 yr), as previously described (Colt et al., 2004). Samples of 4 g of sieved dust were analyzed for the 17 (7 PCDD and 10 PCDF) EPA-designated toxic congeners with high resolution gas chromatograph/high resolution mass spectrometer (HRGC/HRMS) operated in positive electron ionization mode at 10,000 mass resolution, following EPA Method 8290 (U.S.EPA, 2007). The primary analytical column was a J&W 60 m × 0.32 mm I.D. DB-5

column with a 0.25  $\mu$ m film thickness (Agilent, Santa Clara, CA, USA). A secondary confirmation column (for separation of 2,3,7,8-TCDF from its closest eluting isomers) was a J&W 30 m × 0.32 mm I.D. DB-225 column with a 0.25  $\mu$ m film thickness (Agilent, Santa Clara, CA, USA). For concentrations of target analytes with co-eluting interferences, we assumed the interfering compounds (i.e., those inflating the ratio between the two ions monitored for calculation of congener concentrations) affected only one of the monitored ions. The target analyte concentration was then estimated based on the assumed non-interfered ion and the expected ratio between the two.

Quality control samples in each batch of 20 samples included a solvent blank, matrix spike, and matrix spike duplicate.. The median relative percent difference of the matrix spikes and duplicates was 6% with a range of (1%, 52%). A few samples that required dilution due to the very high concentrations of certain PCDD/F congeners had relative percent differences above the acceptable limit of 35% designated by EPA Method 8290. Mean recoveries ranged from 67% to 85%, depending on the congener, with the higher chlorinated compounds having higher recoveries.

#### 2.8. Statistical Analysis

Using a method reported by Lubin et al. (2004), we imputed values below the limit of detection (LOD) by assigning a value for each missing measurement from a lognormal distribution using calculated maximum likelihood parameter estimates using SAS software (SAS Institute, Inc., Cary, NC). Because 1,2,3,7,8,9-HxCDF had a detection rate below 50%, it was excluded from statistical analyses.

We calculated the TEQ in house dust using both the 1998 and 2005 World Health Organization TEFs. We present only the 2005 TEQs for the house dust, because they were highly correlated with the 1998 TEQs (Spearman r=0.996, p<0.0001) and reflect the most up-to-date toxicity information (U.S.EPA, 2010; Van den Berg et al., 2006).

The relative concentration of the PCDD/F congeners (called the "congener profile") can be indicative of specific sources. We derived the congener profile by dividing the concentration of each congener by the sum of the concentrations of the 17 toxic congeners and multiplying by 100 to calculate each congener's contribution to the total toxic PCDD/F concentration (Lorber et al., 1998).

Using separate univariate linear regression models, we evaluated the association between each of the potential determinant and the concentration of each of the 17 toxic congeners and the TEQ. We used the natural logarithm of the PCDD/F concentrations in dust as the outcome variables for all analyses, because they were distributed approximately lognormally. Determinants with p-values of 0.2 or less from the univariate models were considered candidates for inclusion in multivariate models. For each PCDD/F congener and the TEQ, a final, multivariate model was created by first including candidate facility- and traffic-related determinants. Next, candidate demographic and residential variables were added to models to evaluate whether they explained any additional variance in the PCDD/F dust concentrations. Variables with p-values<0.1 were retained in the final models. To account for the additional variance from the imputation of values below the detection limit, we fit each congener's final regression model five times with five imputed datasets and combined the results using the MIANALYZE procedure in SAS (Lubin et al., 2004; Rubin and Schenker, 1991). We additionally evaluated whether the determinants of PCDD/F were also predictors of the five PCB congeners measured in the dust samples (PCB 105, 138, 153, 170, and 180), because PCBs can be emitted by many of the same facility types as PCDD/F (U.S.EPA, 2006) and could be correlated with PCDD/F.

# 3. Results

#### 3.1. Study Population

The demographic and residential characteristics of the 40 pilot study participants and the 1,106 participants of the main study who provided vacuum bags are presented in Table 1. The two populations were similar with respect to case status, sex, smoking status, and whether they ever worked in the refuse industry. There were some differences with respect to study center, with Los Angeles County being under-represented in the pilot population (8%) compared to the larger population (25%), and Iowa being over-represented in the pilot (45%) compared to the larger population (29%). Additionally, in the pilot, 1% of participants had ever been a farmer, while in the larger population 10% had ever been farmers.

Because our pilot study population was originally selected based on the AEI (AEI=0 or AEI>0), rather than proximity to any individual facility type, only 4 individual facility types were located within 5 km of a study home (Table 1). The most common facility type within 5 km of the residences was medical waste incinerator in both the pilot (45%) and the larger study population (24%). In the pilot study, cement kilns, coal-fired power plants, and sewage waste incinerators each had two (5%) different homes within 5 km. The two homes located within 5 km of a cement kiln were each located near the same two cement kilns. No homes in the entire NCI SEER NHL Study were located within 5 km of cement kilns burning hazardous waste, copper smelters, industrial boilers, or iron ore sintering plants (Pronk et al., 2009), and in the pilot study, no homes were located within 5 km of hazardous waste incinerators or municipal solid waste incinerators. The pilot study homes appeared to be a little older than those in the larger population, while the population density and proximity to major roads and freight routes were similar in the two populations.

#### 3.2. Distributions of PCDD/F in House Dust

We found measurable PCDD/F in house dust in all homes. All congeners except 1,2,3,7,8,9-HxCDF were present at levels above the LOD in at least 80% of samples (Table 2). The most toxic congeners, TCDD and 1,2,3,7,8-PeCDD (TEFs=1), had median concentrations of 0.49 pg/g (IQR: 0.30, 0.84) and 2.0 pg/g (IQR: 1.1, 2.8), respectively, and contributed 0.01% and 0.03% to the TEQ. The most abundant congeners were OCDD (TEF=0.0003, median concentration=6000 pg/g, IQR: 3400, 9000), 1,2,3,4,6,7,8-HpCDD (TEF=0.01, median=660 pg/g, IQR: 400, 1090), and OCDF (TEF=0.003, median=280 pg/g, IQR: 140, 500), contributing 84%, 9.2%, and 3.7% to the TEQ. The median TEQ was 20 pg/g (IQR: 14, 33).

# 3.3. Evaluation of Determinants of PCDD/F Concentrations

Estimates of the proportional increase in PCDD/F concentrations in dust with respect to the determinants from the final multivariate models (p<0.1) are presented in Tables 3 and 4. Summary statistics for concentrations with respect to some of the key determinants are reported in Supplemental Table S.1 Residences within 5 km of a cement kiln or sewage incinerator had 2.5- and 2.4-fold higher 2,3,7,8-TCDD dust concentrations, respectively, than residences further away (p<0.05 for both). Residence within 3 km or 5 km of a cement kiln was associated with 2- to 9-fold higher dust concentrations of four other PCDD and five PCDFs (p<0.1 in each model) and a 5-fold higher dust TEQ (p<0.05). Proximity to freight routes was associated with elevated concentrations of one PCDD and five PCDF (p<0.1 in each model), but not any PCDD. The regression coefficients for the traffic-related variables ranged from 0.30 to 0.67, indicating that the concentrations of PCDD/F decreased by approximately one-half for each km distance between a residence and

a freight route or major road. Population density was inversely associated with four PCDD, four PCDF, and the TEQ. The AEI was not associated with concentrations of any PCDD/F. The models explained 10 to 49% of the variability in the PCDD/F concentrations. Dust concentrations of each of the 5 PCBs were elevated in homes located within 5 km of cement kilns, but were not associated with proximity to traffic- or other facility-related sources (not shown). Because PCBs were previously measured in homes that met the dust sampling criteria (n=1106), we were able to determine that there was no relationship between PCB concentrations and proximity to cement kilns within the larger study population (not shown).

# 4. Discussion

To our knowledge, this is the first study to examine multiple outdoor determinants of concentrations of PCDD/F in house dust in different sites across the United States. PCDD/Fs were universally detectable in house dust, and we observed relationships between various PCDD/Fs and certain sources. Proximity to cement kilns was associated with concentrations of several PCDD/Fs. Proximity to freight routes and major roadways was also associated with higher concentrations of numerous compounds, mainly PCDF.

Few studies have evaluated determinants of PCDD/F in house dust. The University of Michigan Dioxin Exposure Study (UMDES) identified residential soil concentrations of PCDD/F as a statistically significant predictor of concentrations of several PCDD/F and the TEQ in residential dust samples in Michigan. In addition, the age of the floor surface was a predictor of nearly all congeners measured, and trash burning at the house was a significant predictor of a few PCDD/F congeners and the TEQ (Jiang et al. 2011; Knutson et al. 2007a; Knutson et al. 2007b).

The median house dust concentration of PCDD/F in our study homes (20 pg TEQ/g) was slightly lower than in homes located downwind of a former incinerator and pesticide producer (27 pg TEQ/g), but slightly higher than a reference population over 100 miles away (11 pg TEQ/g) (UMDES, 2008; Table 5). The median concentrations measured in attic dust samples in four studies of homes within 2 miles of former PCDD/F-releasing facilities were 4 to 20 times higher than what we observed (Table 5). However, in our study, the median TEQ in dust of the two homes located within 5 km (3.1 miles) of cement kilns was 66 pg TEQ/g, which is closer to the concentrations reported in these four studies. The higher concentrations observed in the attic dust studies could be because attic dust may integrate over a longer time period than vacuum bag dust, capturing higher, historical exposures (Ilacqua et al., 2003). There are no health-based standards for PCDD/F concentrations in indoor dust with which to compare concentrations measured in our study.

Proximity to cement kilns was associated with concentrations of most PCDD/Fs that we evaluated. With the exception of TCDD and sewage incinerators, we did not observe associations with other facility types. Our finding that PCDD/F levels are elevated in dust sampled from homes located near cement kilns suggests that PCDD/F might be the putative exposures for the observed elevated risk of NHL among NCI-SEER study participants living within 3 km of a cement kiln (OR=3.8, 95% CI 1.1–14.0) (Pronk et al., submitted). However, results of both these analyses are based on small numbers and need to be explored in a larger number of homes. Further, the pilot study homes located within 5 km of cement kilns also had elevated levels of PCBs in the dust, and therefore co-exposures should be considered in epidemiologic analyses.

We did not observe an association between PCDD/F in dust and proximity to medical waste incinerators, the most prevalent facility in our population. Most likely this is because these incinerators are typically small in terms of volume of waste, and thus have low dioxin emissions on a per facility basis (e.g., average emission level of 0.42 ng TEQ/yr in 1995)

compared to other facility types such as municipal solid waste incinerators (e.g., average emission level of 19.88 ng TEQ/yr in 1995) (EPA, 2006; Pronk et al., submitted). Because of the lack of association with medical waste incinerators and the small number of other facility types in our study, it is not surprising that we did not observe a statistically significant association with the AEI, which combines emissions data across all facilities weighted by the TEQ. Because there was a statistically significant association found between the AEI and a subtype of NHL in a previous analysis (Pronk et al, submitted), expanded analyses to validate the index are warranted.

Proximity to major roads and freight routes was associated with higher concentrations of several congeners, mainly PCDF. The emissions of PCDD/F from gasoline and diesel-powered vehicles are well documented (U.S.EPA, 2006). Two studies have observed elevated concentrations of PCDD/F in samples of soil immediately adjacent to highways, with dramatic declines in concentrations at a distance of 20 m (Benfenati et al., 1992; Sidlova et al., 2009). The contribution of traffic to total PCDD/F exposure should be explored further. Because PCDD/F can be emitted from numerous sources, additional sources of vehicle exhaust could be considered, such as freight loading areas, large parking lots, rail lines, rail terminals, and airports.

We observed an inverse association between concentrations of PCDD/F in house dust and population density. Some studies have reported direct associations between population density and PCDD/F exposure, suggesting that urban and industrial areas are larger sources of dioxins than rural areas (Venier et al., 2009; Viel et al., 2008b; Cleverly et al., 2007). However, a study in Japan also observed an inverse correlation between population density and PCDD/F emissions from incinerator plants, even though the plants in the more densely populated areas were large, high-capacity facilities. The authors attributed this inverse relationship to the better air pollution control technologies of incineration plants in urban areas (Fukuda et al., 2003). This is a possible explanation for the inverse association in our study. Additional explanations could be the higher prevalence of rural dioxin sources such as backyard trash burning, agricultural burning (US EPA 2006; Jiang et al., 2011; Knutson et al., 2007a; Knutson et al., 2007b), and exposure from historical use of pesticides contaminated with PCDD/F (e.g., phenoxyherbicides). Support for the third explanation comes from a previous analysis within the NCI-NHL Study, which found that having a farm-related occupation was significantly associated with higher plasma concentrations of several dioxins and furans (De Roos et al., 2005).

Home age has been suggested as a potential factor contributing to PCDD/F in dust (Franzblau et al., 2009) and has been linked to elevated concentrations of the structurally similar PCBs (Colt et al., 2005). We had speculated that older homes would have higher PCDD/F concentrations due to higher emissions from outdoor sources prior to the widespread adoption of air pollution control technologies (U.S. EPA, 2006). Although the years in which the study homes were built covered a wide range (1851 to 1985), we did not observe a relationship between home age and PCDD/F dust concentrations.

The congener profiles we observed were similar across all homes at all four sites and could not be distinguished from a non-specific, background exposure profile (Cleverly et al., 2007; U.S.EPA, 2006). This is consistent with other studies showing that beyond the immediate vicinity of a source, the congener profile becomes less distinct (Lorber et al., 1998; Ames et al., 2012).

Although we observed some intriguing relationships between industrial facility- and trafficrelated sources and concentrations of PCDD/F in house dust, these associations were limited by a small study population. Few homes were located near certain facilities, such as cement

kilns, coal-fired electricity plants, and sewage waste incinerators. Another limitation was the lack of complete data on other potential sources of dioxins in house dust, such as cigarette smoking (Lofroth and Zebuhr, 1992; Wilson et al., 2008) and outdoor trash burning (Jiang et al., 2011; Knutson et al., 2007a; Knutson et al., 2007b). Despite these limitations, our models based on environmental sources of dioxin emissions explained a substantial portion (10 to 49%) of the variability in the PCDD/F concentrations in residential dust, similar to the regression models in the UMDES (14 to 40%) (Jiang et al., 2011).

Higher concentrations of certain PCDD/F in homes near cement kilns, freight routes, and major roads suggest that these outdoor sources are contributing to indoor environmental exposures. Because these findings were based on a small study population and a small number of facilities, additional analyses based on a larger number of homes are needed to improve our understanding of these relationships.

# Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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# Abbreviations and definitions

| AEI  | average emission index                       |
|------|--|
| EPA  | Environmental Protection Agency              |
| GIS  | geographic information system                |
| GPS  | global positioning system                    |
| IARC | International Agency for Research on Cancer  |
| IQR  | inter-quartile range                         |
| LOD  | limit of detection                           |
| NCI  | National Cancer Institute                    |
| NHL  | non-Hodgkin lymphoma                         |
| PCDD | polychlorinated dibenzo-p-dioxins            |
| PCDF | polychlorinated dibenzofurans                |
| SEER | Surveillance, Epidemiology and End Results   |
| TCDD | 2,3,7,8-tetrachlorodibenzo- <i>p</i> -dioxin |
| TEF  | Toxic Equivalence Factor                     |
| TEQ  | Toxic Equivalence                            |

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# Highlights

- We assessed house dust concentrations of PCDD/F and proximity to outdoor sources.
- We observed higher concentrations of PCDD/F in homes near cement kilns and roads.
- Further study of the contribution of these sources to PCDD/F exposure is warranted.

#### Table 1

#### Demographic and residential characteristics.

|  | Pilot Study Dust | Analysis (n=40) | Parent NHL S | tudy (n=1106) <sup>a</sup> |
|--|------------------|-----------------|--------------|----------------------------|
| <u>Characteristic</u>                                | Numbo            | er (%)          | Numb         | <u>er (%)</u>              |
| Study Center   |                  |                 |              |                            |
| Detroit  | 9 (2             | 23)             | 179          | (16)                       |
| Iowa   | 18 (             | 45)             | 323          | (29)                       |
| LA   | 3 (              | 8)              | 276          | (25)                       |
| Seattle  | 10 (             | 25)             | 328          | (30)                       |
| Case   | 19 (             | 48)             | 637          | (57)                       |
| Male   | 20 (             | 50)             | 591          | (53)                       |
| White  | 38 (             | 95)             | 972          | (88)                       |
| Smoking Status                                       |                  |                 |              |                            |
| Never  | 7 (1             | 18)             | 212          | (19)                       |
| Former   | 6 (1             | 15)             | 169          | (15)                       |
| Current  | 5 (1             | 13)             | 57           | (5)                        |
| Not ascertained                                      | 22 (             | 55)             | 668          | (60)                       |
| Ever Worked in Mills, Refineries, or Refuse Industry | 1 (              | 3)              | 23           | (2)                        |
| Ever Worked in Farming                               | 3 (              | 1)              | 113          | (10)                       |
|  | Within 3 km      | Within 5 km     | Within 3 km  | Within 5 km                |
| <i>Facilities<sup>b,c</sup></i>                      |                  |                 |              |                            |
| Non-hazardous waste cement kilns                     | 1 (3)            | 2 (5)           | 5 (0.5)      | 17 (2)                     |
| Coal fired power plants                              | 0 (0)            | 2 (5)           | 11 (1)       | 35 (3)                     |
| Sewage sludge incinerators                           | 0 (0)            | 2 (5)           | 8 (0.7)      | 31 (3)                     |
| Medical waste incinerators                           | 9 (23)           | 18 (45)         | 130 (12)     | 263 (24)                   |
| Municipal solid waste incinerators                   | 0 (0)            | 0 (0)           | 2 (0.2)      | 18 (2)                     |
| Hazardous waste incinerators                         | 0 (0)            | 0 (0)           | 1 (0.1)      | 11 (1)                     |
| Dioxin emission index >0                             | 21 (             | 52)             | 307          | (28)                       |
|  | Median           | <u>I (IQR)</u>  | Media        | <u>n (IQR)</u>             |
| Census Block Population Density (ppsm) <sup>d</sup>  | 3377 (30         | 8, 5500)        | 3728 (68     | 32, 6897)                  |
| Year Residence Built                                 | 1953 (193        | 31 (1967)       | 1960 (194    | 41 (1976)                  |
| Distance to Freight Route (meters) <sup>e</sup>      | 387 (12          | 9, 675)         | 477 (20      | 8, 1000)                   |
| Distance to Major Road (meters) <sup>e</sup>         | 698 (245         | 5, 1000)        | 944 (37      | 8, 1000)                   |

<sup>a</sup>Number of participants from the NCI SEER NHL Study who provided a dust sample, though only the 40 pilot samples were analyzed.

<sup>b</sup>No homes in entire NCI SEER NHL Study were located within 5 km of cement kilns burning hazardous waste, copper smelters, industrial boilers, or iron ore sintering plants

<sup>C</sup>The # of unique facilities within 5 km of homes in the pilot population were: non-hazardous waste cement kilns: 2; coal-fired power plants: 4; sewage sludge incinerators: 2; medical waste incinerators: 28.

<sup>d</sup>2000 U.S. Census of Population and Housing

 $e^{e}$ Residences more than 1000 m from a freight route or major road were assigned a distance of 1000 m.

| Congener                          | TEF    | Median LOD | % Detected | Median Concentration <sup>a</sup> | Inter-Quartile Range | % of Total PCDD/F <sup>b</sup> |
|-----------------------------------|--------|------------|------------|-----------------------------------|----------------------|--------------------------------|
| <u>PCDD</u>                       |        |            |            |                                   |                      |                                |
| 2,3,7,8-TCDD                      | -      | 0.4        | 83         | 0.49                              | (0.28, 0.84)         | 0.01                           |
| 1,2,3,7,8-PeCDD                   | 1      | 1.0        | 93         | 2.0                               | (1.1, 2.8)           | 0.03                           |
| 1,2,3,4,7,8-HxCDD                 | 0.1    | 1.0        | 06         | 4.0                               | (2.2, 6.2)           | 0.05                           |
| 1,2,3,6,7,8-HxCDD                 | 0.1    | 1.0        | 100        | 18                                | (13, 35)             | 0.26                           |
| 1,2,3,7,8,9-HxCDD                 | 0.1    | 1.0        | 100        | 12                                | (8.7, 18)            | 0.16                           |
| 1,2,3,4,6,7,8-HpCDD               | 0.01   | 1.0        | 100        | 660                               | (400, 1090)          | 9.2                            |
| OCDD                              | 0.0003 | 2.0        | 100        | 6000                              | (3400, 9000)         | 84                             |
| PCDF                              |        |            |            |                                   |                      |                                |
| 2,3,7,8-TCDF                      | 0.1    | 0.4        | 100        | 2.1                               | (1.6, 2.8)           | 0.03                           |
| 1,2,3,7,8-PeCDF                   | 0.03   | 1.0        | 98         | 1.4                               | (1.0, 2.2)           | 0.02                           |
| 2,3,4,7,8-PeCDF                   | 0.3    | 1.0        | 98         | 2.4                               | (1.6, 3.8)           | 0.03                           |
| 1,2,3,4,7,8-HxCDF                 | 0.1    | 1.0        | 98         | 8.1                               | (4.4, 15)            | 0.11                           |
| 1,2,3,6,7,8-HxCDF                 | 0.1    | 1.0        | 100        | 5.6                               | (3.0, 9.4)           | 0.07                           |
| $1,2,3,7,8,9-HxCDF^{\mathcal{C}}$ | 0.1    | 1.0        | 48         | 0.67                              | (0.30, 0.70)         | 0.01                           |
| 2,3,4,6,7,8-HxCDF                 | 0.1    | 1.0        | 98         | 5.7                               | (3.1, 11)            | 0.08                           |
| 1,2,3,4,6,7,8-HpCDF               | 0.01   | 1.0        | 100        | 110                               | (59, 280)            | 1.4                            |
| 1,2,3,4,7,8,9-HpCDF               | 0.01   | 1.0        | 93         | 7.0                               | (3.2, 15)            | 0.0                            |
| OCDF                              | 0.003  | 2.0        | 100        | 280                               | (140, 500)           | 3.7                            |
| $TEQ^d$                           |        |            |            | 20                                | (14, 33)             |                                |

Distributions of PCDD/F concentrations in house dust  $(pg/g)^{a}(n=40)$ 

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 $^{b}$ Calculated as the congener concentration divided by the concentration of all 17 toxic congers imes 100%.

 $^{d}\!\mathrm{TEQ}$  calculated using the 2005 World Health Organization toxic equivalency factors.

 $\boldsymbol{\mathcal{C}}_{\text{Excluded}}$  from further analyses due to low detection.

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# Table 3

Proportional increase in PCDD concentrations in house dust associated with proximity to dioxin sources, multivariate regression models.

| PCDD Congener<br>Determinant   | exp(b) (95% Confidence Interval) | R-squared <sup>a</sup> |
|--------------------------------|----------------------------------|------------------------|
| 2,3,7,8-TCDD                   |                                  |                        |
| Cement Kiln 5 km               | 2.5 (1.1, 5.6) **                | 0.24                   |
| Sewage Incinerator 5 km        | 2.4 (1.1, 5.5) **                |                        |
| 1,2,3,7,8-PeCDD                |                                  |                        |
| Distance to Freight Route (km) | 0.57 (0.33, 0.97)**              | 0.15                   |
| Population Density (1000 ppsm) | 0.96 (0.93, 1.00) **             |                        |
| 1,2,3,4,7,8-HxCDD <sup>b</sup> |                                  |                        |
| Cement Kiln 3 km               | 7.6 (1.2, 47)**                  | 0.23                   |
| Population Density (1000 ppsm) | 0.96 (0.91, 1.0)*                |                        |
| 1,2,3,6,7,8-HxCDD              |                                  |                        |
| Cement Kiln 3 km               | 6.7 (0.92, 49)*                  | 0.17                   |
| Population Density (1000 ppsm) | 0.55 (0.89, 1.0)*                |                        |
| 1,2,3,7,8,9-HxCDD              |                                  |                        |
| Cement Kiln 3 km               | 5.7 (1.2, 28)**                  | 0.20                   |
| Population Density (1000 ppsm) | 0.95 (0.91, 0.99)**              |                        |
| 1,2,3,4,6,7,8-HpCDD            |                                  |                        |
| Cement Kiln 3 km               | 8.6 (1.06, 69)**                 | 0.10                   |
| OCDD                           |                                  |                        |
| None                           | NA                               | NA                     |

\* p<0.10,

\*\* p<0.05

 $^{a}$ The r-squared statistic was derived from final regression model with the first imputed value.

<sup>b</sup>adjusted for gender

PCDD, polychlorinated dibenzo-p-dioxins; ppsm, people per square mile; NA, not applicable

# Table 4

Proportional increase in PCDF and TEQ concentrations in house dust associated with proximity to dioxin sources, multivariate regression models.

| PCDF Congener<br>Determinant   | exp(b) (95% Confidence Interval) | R-squared <sup>a</sup> |
|--------------------------------|----------------------------------|------------------------|
| 2,3,7,8-TCDF <sup>b</sup>      |                                  |                        |
| Cement Kiln 5 km               | 3.1 (1.7, 5.9) **                | 0.50                   |
| Distance to Freight Route (km) | 0.67 (.47, 1.0)*                 |                        |
| 1,2,3,7,8-PeCDF <sup>b</sup>   |                                  |                        |
| Cement Kiln 5 km               | 3.7 (1.6, 8.7) **                | 0.49                   |
| Distance to Freight Route (km) | 0.60 (0.37, 1.0)*                |                        |
| 2,3,4,7,8-PeCDF <sup>b</sup>   |                                  |                        |
| Cement Kiln 5 km               | 3.9 (1.7, 9.0) **                | 0.42                   |
| Distance to Freight Route (km) | 0.48 (0.30, 0.79)***             |                        |
| 1,2,3,4,7,8-HxCDF              |                                  |                        |
| Distance to Freight Route (km) | 0.30 (0.14, 0.65) **             | 0.24                   |
| Population Density (1000 ppsm) | 0.95 (0.90, 1.0) **              |                        |
| 1,2,3,6,7,8-HxCDF              |                                  |                        |
| Cement Kiln 5 km               | 2.8 (1.1, 7.9) **                | 0.33                   |
| Distance to Major Road (km)    | 0.41 (0.24, 0.71)**              |                        |
| Population Density (1000 ppsm) | 0.96 (0.93, 1.0)*                |                        |
| 2,3,4,6,7,8-HxCDF              |                                  | 0.22                   |
| Cement Kiln 5 km               | 3.0 (1.1, 8.1) **                |                        |
| Distance to Major Road (km)    | 0.48 (0.27, 0.84)**              |                        |
| 1,2,3,4,6,7,8-HpCDF            |                                  | 0.16                   |
| Distance to Freight Route      | 0.47 (0.19, 1.2)*                |                        |
| Population Density (1000 ppsm) | 0.94 (0.88, 0.99) **             |                        |
| 1,2,3,4,7,8,9-HpCDF            |                                  |                        |
| Population Density (1000 ppsm) | 0.93 (0.87, 0.99)**              | 0.11                   |
| OCDF                           |                                  |                        |
| Distance to Major Road (km)    | 0.45 (0.21, 0.78) **             | 0.10                   |
| TEQ                            |                                  |                        |
| Cement Kiln 3 km               | 5.3 (1.0, 28)**                  | 0.16                   |
| Population Density (1000 ppsm) | 0.96 (0.91, 1.0) **              |                        |

\* p<0.10,

\*\* p<0.05

 $^{a}\!\!\!\mathrm{The}$  r-squared statistic was derived from final regression model with the first imputed value.

<sup>b</sup>adjusted for Study Center

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PCDF polychlorinated dibenzofurans; ppsm, people per square mile; TEQ, toxic equivalence

| Sample Location (Reference)  | Z   | Median (pg TEQ/g) <sup>a</sup> | Range (pg TEQ/g) <sup>d</sup> | Study Period | Dust Sample Source                                 |
|--|-----|--------------------------------|-------------------------------|--------------|--|
| LA, Detroit, Seattle, Iowa (current study)   |     |                                |                               |              |  |
| general U.S. population  | 40  | 20                             | (5.4, 260)                    | 1998–1999    | vacuum bag   |
| Michigan (UMDES 2008)  |     |                                |                               |              |  |
| Jackson/Calhoun (reference pop; >100 miles from former incinerator/<br>pesticide producer) | 198 | 11                             | (1.6, 1060)                   | 2004–2005    | hard and carpeted flooring in frequently used room |
| Midland plume (downwind former incinerator/pesticide producer) $b$                         | 37  | 27                             | (7.6, 96)                     | 2004–2005    |  |
| Southern Alabama (Hensley 2007)  |     |                                |                               |              |  |
| within 1 mile of former wood treatment facility  | 11  | 84                             | (8.4, 502)                    | 2006         | attic  |
| Mississippi (Dahlgren 2007)  |     |                                |                               |              |  |
| within 2 miles of former wood treatment facility   | 38  | 75                             | (0.91, 22000)                 | 2007         | attic  |
| Sauget, Illinois (Gonzalez 2011)   |     |                                |                               |              |  |
| within 2 miles of former PCB/pesticide producer & secondary copper facilities              | 14  | 348                            | (29, 140000)                  | 2008         | attic  |
| Louisiana, Mississippi, Alabama (Feng 2011)  |     |                                |                               |              |  |
| within 2 miles of former wood treatment facilities   | 60  | 139                            | (8.2, 13936)                  | not provided | attic  |

<sup>a</sup>TEQs weighted by WHO 2005 TEFs.

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 $\boldsymbol{b}_{\text{Based}}$  on a erosol dispersion model; distances from source not provided.

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Table 5