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# Residential proximity to agricultural herbicide and fungicide applications and dust levels in homes of California children

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## Abstract

**Background:** Few studies of the relationship between residential proximity to agricultural pesticide applications and pesticide levels in the home have incorporated crop location or wind direction. We evaluated the relationship between agricultural pesticide applications using the California Pesticide Use Reporting (CPUR) database and pesticide concentrations in carpet dust accounting for land use and wind direction.

**Methods:** We measured concentrations (ng/g) of seven herbicides and two fungicides in carpet dust samples from 578 California homes (2001–2007). We created three metrics by computing the density (kg/km$^2$) of use of each pesticide reported in CPUR within 0.5-, 1-, 2-, and 4-km buffers around homes 180- and 365-days before sampling (CPUR metric). We apportioned applications

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Declaration of competing interest
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement
**Jessica M. Madrigal:** Writing – review & editing, Writing – original draft, Software, Methodology, Formal analysis, Data curation, Conceptualization. **Robert B. Gunier:** Writing – review & editing, Methodology, Investigation, Conceptualization. **Rena R. Jones:** Writing – review & editing. **Abigail Flory:** Writing – review & editing, Software, Data curation. **Catherine Metayer:** Writing – review & editing, Funding acquisition, Data curation, Conceptualization. **John R. Nuckols:** Writing – review & editing, Conceptualization. **Mary H. Ward:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Appendix A. Supplementary data
Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2024.109024.

to the crop area within the buffers (CROP-A metric) and weighted CPUR applications by the proportion of days that the home was within ±45° of the downwind direction (W-CPUR metric). We modeled natural-log concentrations (Tobit regression) and dust detections (logistic regression) adjusting for season/year, occupation, and home/garden use.

**Results:** Detections were >90 % for glyphosate, 2,4-D, and simazine. Detection rates and dust concentrations increased with increasing CPUR densities for all herbicides and one fungicide. Compared to homes without applications within 4 km, the highest tertile of 365-day glyphosate use was associated with ~100 % higher concentrations ($\mathrm{CPUR}_{\mathrm{T3} > 9.2\mathrm{kg/km}^2}\%_{\mathrm{change}} = 110,\ 95\ \%\mathrm{CI} = 55,\ 183$; $\mathrm{CROP} - \mathrm{A}_{\mathrm{T3} > 13.4\mathrm{kg/km}^2}\%_{\mathrm{change}} = 144,\ 95\ \%\mathrm{CI} = 81,\ 229$; and $\mathrm{W} - \mathrm{CPUR}_{\mathrm{T3} > 2.1\mathrm{kg/km}^2}\%_{\mathrm{change}} = 102,\ 95\ \%\mathrm{CI} = 50,\ 171$). The highest density tertiles of 2,4-D, simazine, and trifluralin were associated with 2- to 6-times higher concentrations, respectively; that was similar across metrics. Across all metrics, agricultural use of dacthal, dicamba, and iprodione were associated with 5- to 10-times higher odds of dust detections. Associations were unclear for 2-methyl-4-chlorophenoxyacetic acid and null for chlorothalonil.

**Conclusions:** Agricultural herbicide and fungicide use was an important determinant of indoor contamination within 4 km of homes. Accounting for crops and wind direction did not substantially change these relationships.

**Keywords**

Pesticides; Herbicides; Glyphosate; Fungicides; Agriculture; GIS; Environmental exposure assessment; Dust

## 1. Introduction

Agricultural pesticide use has been increasing globally with an estimated 2.7 million tons of active ingredients used in 2020 (FAO, 2022). In the United States (US), herbicides account for nearly 60 % of these pesticides (Atwood and Paisley-Jones, 2017; Benbrook, 2016). The herbicide glyphosate is the most highly used agricultural pesticide in the US and globally (Benbrook, 2016) and is the second most used in homes and gardens in the US (Atwood and Paisley-Jones, 2017). Pesticides applied to agricultural fields travel beyond the treated area potentially impacting the health of surrounding residents (Zivan et al., 2017; Dereumeaux et al., 2020; Teysseire, 2021). Children may be particularly vulnerable to pesticide toxicities due to their greater exposure through hand-to-mouth behaviors, higher respiratory rates, higher ratio of body surface area to volume, and closer proximity to the ground. Identifying populations who may be at increased risk for exposure through the proximity of their residences to pesticide applications and understanding factors that influence primary pesticide drift (i.e., the movement of a pesticide away from the intended target area during application as a droplet, vapor or aerosol) and secondary pesticide drift (i.e., the movement of a pesticide after application via evaporation, vapor drift, wind erosion, etc.) are critical to studying health effects associated with exposure to agricultural pesticides.

Numerous epidemiologic studies have evaluated residential proximity to agricultural pesticide applications and childhood cancer (Reynolds, 2005; Park, 2020; Patel, 2020; Lombardi, 2021; Gómez-Barroso, 2016; Thompson et al., 2022; Carozza et al., 2008;

Carozza, 2009) and other pediatric health outcomes, including cognitive function and neurodevelopment (Coker, 2017; Gunier, 2017; Rowe, 2016; Shelton, 2014; von Ehrenstein, 2019; Gunier, 2017; Hyland, 2021). Several of these studies found positive associations between proximity-based exposure metrics and risk of childhood leukemia (Park, 2020; Patel, 2020; Gómez-Barroso, 2016), central nervous system tumors (Lombardi, 2021), retinoblastoma (Thompson et al., 2022) as well as intelligence quotient (Gunier, 2017; Rowe, 2016), autism spectrum disorder (Shelton, 2014; von Ehrenstein, 2019), and depression (Hyland, 2021). These spatial metrics are proxies for exposure and their design varies by study; information used to create these metrics can include residential distance to agricultural fields, crop density around the home, and density of agricultural pesticides applied near the home at various distances. In California, many studies used the state's unique pesticide use reporting (CPUR) database maintained by the California Department of Pesticide Regulation. The CPUR pesticide use data are reported by crop type for each public land survey system section (PLSS; approximately one square-mile) but lack specificity of the exact location of the pesticide application. Only a few studies (Harnly, 2009; Gunier, 2014; Chevrier, 2014) have incorporated ancillary information on crop field locations or wind direction into their CPUR-based exposure metrics. Furthermore, there have been few exposure studies to validate these proximity-based metrics (Harnly, 2009; Gunier, 2011; Madrigal, 2023; Deziel, 2017) by comparing them to objectively assessed exposures, such as concentrations of pesticide active ingredients in house dust or biological samples. Further, only a small number of the exposure studies evaluated the timing of pesticide applications, varying distances of applications from the home, or meteorological conditions, and these have been limited to a few pesticide active ingredients. To date, no study has evaluated the relationship between residential proximity-based metrics of glyphosate use and levels in house dust in the US.

Previously, we used the CPUR database (California Department of Pesticide Regulation, 2020) to create exposure metrics for nine insecticides that incorporated crop location, wind direction, and varying distances and time periods and showed that agricultural use of six of the nine insecticides within 4 km of homes predicted house dust concentrations (Madrigal, 2023). We demonstrated the importance of creating metrics that were tailored to each active ingredient in order to best predict insecticide concentrations in homes. Here, we extend this work to herbicides and fungicides, including glyphosate. Our aim was to assess agricultural herbicide and fungicide use near residences of children and evaluate the importance of distance, timing of applications, application method, location of crops, and wind direction as determinants of concentrations in homes.

## 2. Methods

### 2.1. Study population

The California Childhood Leukemia Study (CCLS) is a population-based case-control study of childhood leukemia in 35 counties in the Central Valley and San Francisco Bay area (Chang, 2006; Ma, 2004; Ma, 2005). In 2001 to 2007, CCLS participants who were <8 years old at diagnosis and who were living in the same home where they resided before diagnosis (comparable reference date for controls) were invited to participate in a second interview in which a carpet dust sample was collected (Ward, 2009; Colt, et al., 2008). Our study included 578 residences from CCLS participants who completed the second interview. The time between diagnosis/reference date and dust sample collection ranged from 0.4 to 4.7 years (median = 1.2; interquartile range 0.9, 1.8). Among those who participated in the second interview, 73 % were from urban, 12 % from suburban, and 13 % from rural areas. The CCLS study protocol was approved by the institutional review boards at the University of California, Berkeley, the California Committee for Protection of Human Subjects, and the National Cancer Institute.

### 2.2. Interviews and dust sampling

Interviewers took a Global Positioning System (GPS) reading outside of the home and asked participants about the household's pesticide use and whether anyone in the home had a pesticide-exposed job in the previous twelve months (Deziel, 2015).

The dust sampling methods have been previously described (Colt, et al., 2008). Briefly, we identified the room in which the child had spent the most time while awake during the year before diagnosis (reference date for controls). If that room had a carpet or area rug measuring at least 0.84 square meters (9 square feet) that was present before diagnosis, we used a high-volume surface sampler vacuum (HVS3; Cascade Stack Sampling System, Venice, FL) to take the sample. For most subjects, the sample room was the living room or family room. Dust was also collected from the household vacuum and if the amount of HVS3 dust was insufficient, we analyzed the vacuum dust. In our analysis, dust from the HVS3 was used for 65 % to 76 % of the samples measured using the three different extraction methods, and the rest came from the household vacuum. A prior exposure study that included a small subset of CCLS homes (n = 33) determined that the household vacuum cleaner method is a reasonable alternative to the HVS3 for detecting, ranking, and quantifying the concentrations of pesticides in carpet dust (Colt, et al., 2008).

### 2.3. Laboratory analysis

Dust samples were shipped to Battelle Memorial Institute (Columbus, OH) where they were stored at −20° Celsius until analysis. We measured eight herbicides (2,4-dichlorophenoxyacetic acid (2,4-D), dicamba, 2-methyl-4-chlorophenoxyacetic acid (MCPA), 2-(4 chloro-2-methylphenoxy) propionic acid (MCPP), chlorthal-dimethyl (dacthal), trifluralin, simazine, and glyphosate) and two fungicides (chlorothalonil and iprodione) that had agricultural use in the study area counties during the study period and could be quantified in dust using valid and reliable extraction and quantification methods. The laboratory quantification methods and quality control procedures have been previously described (Colt, et al., 2008; Metayer, 2013; Ward, 2023). Briefly, analytes except glyphosate were extracted using either a herbicide acid or hexane:acetone extraction method and extracts were quantified using gas chromatography/mass spectrometry (MS). In 2019, the laboratory developed a new method (liquid chromatography tandem MS) for quantification of glyphosate, which was used to measure glyphosate for participants that had at least 2 g of dust remaining. Glyphosate was extracted with 1 % formic acid in methanol, followed by purification by C18 Solid Phase Extraction cartridges with final elution into 1 mL of methanol and then was quantified using a liquid chromatography tandem mass spectrometry method (Ward, 2023). Detection limits ranged from 1 ng/g dust for dacthal to 20 ng/g dust for iprodione, and 47 ng/g (0.047 μg/g) dust for glyphosate. Mean sample recoveries generally ranged from 81 % to 125 %. Analytes with lower recovery means included dicamba (33 ± 20 %), chlorothalonil (48 ± 55 %), and MCPP (69 ± 35 %) while analytes with higher recovery means included iprodione (139 ± 27 %) (Colt, et al., 2008). For glyphosate, mean sample recoveries in the replicate samples ranged from 81 to 118 % with the exception of greater variability in three batches (−26 %, 8.4 %, 255 %) that was likely due to two low recoveries and one high recovery in the spiked samples (Ward, 2023).

## 2.4. Pesticide use metrics

The California Department of Pesticide Regulation has required reporting of all pesticide applications since 1990 for PLSS (California Department of Pesticide Regulation, 2020). The resulting CPUR database (California Department of Pesticide Regulation, 2017) can be used to identify the active ingredient, date and method of application, crop treated, and the PLSS section and acreage treated for all agricultural pesticide applications and some non-agricultural applications (e.g. recreation areas, landscape maintenance). We linked the location of the participant's home (GPS coordinate) to the CPUR data using a GIS [ArcGIS Pro 3.1.2 (Esri, Redlands, CA)] and obtained the pounds applied, application method (aerial or ground spraying), and PLSS area for all applications within 4 km in the 180- and 365-day periods prior to the dust collection. We created metrics for each residence and active ingredient using buffers with radii of 0.5-, 1-, 2-, and 4-km (buffer areas: 0.79 $km^2$, 3.14 $km^2$, 12.57 $km^2$, and 50.27 $km^2$, respectively) for each of two time periods (180 and 365 days before dust collection). We excluded applications to non-agricultural targets, including rights-of-way and recreation areas, which accounted for 0.13 % of the total pesticide use records that matched to participant residences during our study period.

We created three metrics for each active ingredient: 1) a CPUR density metric (Supplemental Fig. 1); 2) a crop-area adjusted metric (CROP-A; Supplemental Fig. 2); and 3) a wind-adjusted CPUR metric (W-CPUR; Supplemental Fig. 3). The methods to create these metrics have been described previously (Madrigal, 2023). In short, the CPUR density metric was the sum of area-weighted kilograms of each pesticide active ingredient applied in PLSSs that intersected a circular buffer around participant's residence divided by the area of the buffer (Gunier, 2011; Madrigal, 2023; Nuckols, 2007).

To identify the location of agricultural land for the CROP-A metric, we used 30 m resolution geographic data on pasture/hay and row crop (e.g., vegetables, orchards, vineyards) locations from the 2001, 2004, and 2006 versions of the National Land Cover Database (NLCD) (Homer, 2007; Xian et al., 2009; Fry, 2011). We matched residence locations to the NLCD product that most closely corresponded to the period prior to the dust collection. We created the crop land use metric by calculating the proportion of agricultural land within the PLSS intersecting the buffer out of the total agricultural land in the PLSS, multiplying it by the amount of pesticide applied within the PLSSs, then summing across all the sections within the buffer, and dividing by the total agricultural land area within the buffer.

The W-CPUR metric accounts for wind conditions on the dates of applications and subsequent days up to the date of dust collection. Daily wind direction was derived from the North American Regional Reanalysis database (spatial resolution of 32 km × 32 km and temporal resolution of 1 day) (Mesinger, et al., 2006). We considered the contributing area for drift to be within a 90° (±45°) downwind ‘capture zone’ wedge centered on the wind direction and anchored at the section centroid. We weighted pesticide use in each section according to the percentage of time the home was downwind during the period between application and dust collection.

## 2.5. Statistical analyses

We examined the number of residences with agricultural herbicide and fungicide applications within the 0.5 km and 4 km buffers for each metric (CPUR, CROP-A, and W-CPUR). We excluded MCPP from further analysis due to limited applications (1 home for the CPUR 0.5 km metric, 17 homes for 4 km). For each of the nine remaining active ingredients, we computed Spearman rank correlations to evaluate the associations between the three metrics for each buffer and time period. We also determined the percentage of applications using ground and aerial spraying.

For active ingredients that had at least a 60 % detection rate (2,4-D, trifluralin, simazine, glyphosate), we assigned dust values below the detection limit using a single imputation assuming a log-normal distribution (Lubin, 2004). Since the pesticide concentrations (ng/g) were not normally distributed, we used non-parametric tests to compare detection frequencies and distributions, overall and among homes with and without agricultural use within 4 km. We used Tobit regression to model the natural log transformed dust concentrations. We report the percent change in dust concentration, 100 %*(exp(β)-1), comparing categories of the CPUR, CROP-A, and W-CPUR metrics to residences with no agricultural pesticide use. For active ingredients detected in <40 % of dust samples (dicamba, MCPA, chlorothalonil, dacthal, iprodione), we used logistic regression to model the odds of the active ingredient being detected in the dust (detect vs. non-detect). For dacthal (detection rate: 35 %) and dicamba (detection rate: 28 %), we also used ordinal logistic regression to model the concentrations split at the median compared to non-detections. We categorized the CPUR, CROP-A, and W-CPUR metrics based on their respective distributions as follows: as indicator variables (any applications vs. none) if agricultural use in the buffer was <10 %; as three categories of none, ≤ median and >median for pesticides with agricultural use ranging from 10–25 %; and as tertiles with a separate no use category for pesticides with agricultural use >25 %. When a pesticide metric was modeled as an indicator variable at a smaller distance but a categorical variable at larger distances, we included estimates for indicator variables at all distances to facilitate comparisons.

We examined a range of covariates using a forward selection approach, including household characteristics known to be predictors of pesticide concentrations in dust (Deziel, 2015). This included binary (yes/no) variables for self-reported pesticide use in and around the home and garden in the previous 12 months. Respondents were queried about treatment of ants/termites, fleas/ticks in home or on pets, bees/wasps/hornets, flies/mosquitos, indoor plants, lawn/garden insects, lawn/garden weeds, professional inside treatments, professional outdoor insect treatments (including to the foundation, exterior, or lawn), and professional treatment of lawn/garden weeds. We evaluated whether household members usually removed their shoes before entering the home, and whether one or more persons in the home had a pesticide-exposed occupation (agricultural packing, farming, and pesticide mixing). We also evaluated year (continuous variable from 2001 to 2006, centered by subtracting 2000), and season (winter, spring, summer, and fall) of dust collection as predictors of dust concentrations. Any covariate with a p-value $\leq 0.1$ was retained in the final multivariable model for that pesticide. For glyphosate, 2,4-D, trifluralin, and simazine (>60 % detections), we assessed the application method as a predictor of dust concentrations in the 0.5 km and 4 km buffers by creating separate CPUR metrics for ground and aerial spraying applications and including them together in the multivariable models for each pesticide. All analyses were conducted in SAS version 9.4 (Cary, NC) and STATA SE version 18.

## 3. Results

Agricultural use of the 8 herbicides and 2 fungicides was widespread in the study counties (Fig. 1). Based on the CPUR metric, glyphosate was applied near the highest proportion of homes (4 km: 67 % of homes), followed by iprodione (41 %) (Table 1). The median density of use for glyphosate ranged from 5.5 $kg/km^2$ to 2.7 $kg/km^2$ in the 0.5 km and 4 km buffers, respectively. The median density of use for the other pesticides was less than 5 $kg/km^2$, except for dacthal, for which it was 12.2 $kg/km^2$ at 0.5 km (1.9 at 4 km).

Prevalence of use within the buffers was similar across the CPUR and W-CPUR metrics, with lower prevalence for the CROP-A metric (Supplemental Table 1). Except for MCPA, which was applied by aerial spraying for 44 % of applications within 4 km, the pesticides were applied predominantly using ground spraying (72.6 % of dicamba to 95 % of glyphosate applications; Supplemental Table 2).

We evaluated the Spearman rank correlations between the density metrics for each buffer and time period (Supplemental Table 3). Correlations for the 180-day period were similar to those for 365-days, except for the CROP-A metrics for MCPA and dicamba, which were lower in the 365-day period relative to the 180-day period for the 0.5 km and 2 km buffers. Correlations between the CPUR, CROP-A, and W-CPUR metrics were otherwise high ($\rho_S > 0.7$) for all the pesticides for both the 2 km and 4 km buffers and for both time periods. Correlations between the metrics generally increased with greater buffer distances. In the 0.5 km buffer, metrics for trifluralin, iprodione, and chlorothalonil were less correlated, especially between the CROP-A and W-CPUR metrics. For example, correlations between CROP-A and W-CPUR for 180- and 365-day trifluralin were both $\rho_S = 0.2$.

Detections of the active ingredients in the dust samples ranged from 12–15 % for iprodione and MCPA, respectively, to >90 % for glyphosate, 2,4-D, and simazine (Table 2). Among homes with herbicide detections, median dust concentrations ranged orders of magnitude from <10 ng/g for trifluralin, dacthal, and dicamba to >100 ng/g for 2,4-D and >1000 ng/g for glyphosate. Median concentrations were higher in homes with nearby agricultural use compared with no use except for dicamba and MCPA that had similar median concentrations. However, detection rates for dicamba and MCPA were higher in homes with agricultural use within 4 km versus no use (38 vs. 26 % and 21 % vs. 13 %, respectively).

For the five herbicides detected in >60 % of homes, median concentrations were higher in homes with nearby agricultural use (Fig. 2) with the largest differences in concentrations for glyphosate and 2,4-D. Among homes with fungicide detections, median dust concentrations were 21 ng/g for chlorothalonil and 48 ng/g for iprodione. Median concentrations were similar in homes with and without nearby agricultural use. Detection rates for iprodione, but not chlorothalonil, were higher in homes with agricultural use within 4 km versus no use (23 % vs. 5 % and 19 % vs. 18 %, respectively).

All three metrics were significant predictors of dust concentrations for the four herbicides detected in more than 60 % of dust samples (glyphosate, simazine, 2,4-D, and trifluralin; Table 3). For each active ingredient, we present the associations for the 365-day metrics in the main results tables (Tables 3 and 4). In multivariable models, glyphosate concentrations were 83 % to 110 % higher in homes with the highest density of use (365-day CPUR metric) compared to homes with no agricultural use in the 0.5 km and 4 km buffers, respectively, with significant linear trends. With the exception of a weaker positive trend for the 0.5 km CROP-A metric, associations were similar using the CROP-A and W-CPUR metrics, including for the 180-day metrics (Supplemental Table 4).

For simazine, the 365-day metrics were associated with ~150–300 % greater concentrations in the 0.5 km-4 km buffers and the linear trend was significant across density categories for all three metrics (Table 3). Similar patterns of associations were observed for the 180-day metrics but increases in dust concentrations across the higher density categories were larger (~200 %-400 %; Supplemental Table 4).

For 2,4-D, we observed increases ranging from 60 % in the 0.5 km buffer to 102 % in the 4 km buffer with significant linear trends for the 365-day CPUR metric (Table 3). Estimates varied somewhat but we observed similar significantly positive trends using the CROP-A and W-CPUR metrics. Similar patterns of associations were observed for the 180-day metrics but increases in dust concentrations across the higher density categories were larger ranging from 147 % in the 0.5 km buffer to 138 % in the 4 km buffer (CPUR metric; Supplemental Table 4).

For trifluralin, the highest density use categories for all three 365-day metrics were associated with 400–600 % higher concentrations compared to homes with no use across all the buffer sizes with significant linear trends (Table 3). Somewhat smaller increases were observed with the 180-day metrics (Supplemental Table 4).

Comparing applications of glyphosate, simazine, 2,4-D, and trifluralin by aerial and ground spraying, we found similar percent increases in herbicide concentrations with increasing application densities for both methods (Supplemental Table 5). Since aerial applications were used less frequently than ground spraying, 0.5 km aerial CPUR metrics for simazine, 2,4-D, and trifluralin could only be categorized as any/none.

For the three herbicides (MCPA, dacthal, and dicamba) and two fungicides (iprodione and chlorothalonil) detected in <50 % of homes, we modeled the odds of detection. For MCPA, the 365-day 4 km CPUR, CROP-A and W-CPUR metrics showed the strongest associations with significant trend with increasing density for the W-CPUR metric (Table 4). In the smaller buffers, confidence intervals were imprecise. Patterns were similar using the 180-day metrics (Supplemental Table 6).

Odds of dacthal detection in relation to metrics of any agricultural use vs. no use (Table 4) ranged from 20-fold higher in the 0.5 km buffer to about six-fold higher in the 4 km buffer for the 365-day CPUR, CROP-A, and W-CPUR metrics with similar patterns for the 180-day metrics (Supplemental Table 6). Odds of dicamba detection were also similar for the 365-day CPUR, CROP-A, and W-CPUR metrics and ranged from 4-fold higher in the 0.5 km buffer to about 2-fold higher in the 4 km buffer, though associations were non-monotonic when we split the 4 km density metric at the median (Table 4). Patterns were similar using the 180-day metrics (Supplemental Table 6) and when we modeled the dust concentration split at the median (Supplemental Table 7).

For iprodione, odds of detection ranged from 5 to 6-fold higher in the 0.5 km buffer to 8 to 10-fold higher in the 4 km buffer for the 365-day CPUR, CROP-A, and W-CPUR metrics with significant linear trend across increasing density categories (Table 4). Associations were strongest for the highest categories of the W-CPUR metric compared with the highest categories of the other metrics for all buffers. We observed similar patterns for the 180-day metrics (Supplemental Table 7). Associations for chlorothalonil were mostly null with wide confidence intervals and no significant trends for both the 365-day (Table 4) and 180-day metrics (Supplemental Table 6).

## 4. Discussion

In this population-based study of families with young children, we found strong associations between our three proximity-based metrics of agricultural pesticide use and house dust concentrations of six of the seven herbicides and one of the two fungicides. The CPUR metrics were generally highly correlated with our metrics adjusted for land use and wind direction. All three metrics showed similar patterns of associations with concentrations of the six herbicides and one fungicide in homes. Our results demonstrate that pesticides travel varying distances and that some may travel at least 4 km from treated fields.

This study builds on prior work by Gunier et al. in this study population that evaluated dacthal, simazine, and iprodione in 89 homes and showed agricultural use of these pesticides within 1.25 km of the residence was a significant determinant of concentrations in house dust (Gunier, 2011). In our study, we included a larger number of homes, evaluated additional distances and additional herbicide and fungicide active ingredients, and incorporated wind direction in the metric. Use of dacthal, simazine, and iprodione within 0.5 km to 4 km in the 180 and 365 days before dust collection was associated with dust concentrations in the home, with sizable increases in dust concentrations (e.g., 200–400 % higher for simazine) comparing homes with the highest density of agricultural use to those that were non-exposed. We had similar findings for other ingredients that were not evaluated in the Gunier et al. study, including glyphosate, 2,4-D, and trifluralin. Associations were strongest for trifluralin with concentrations in dust that were 500–600 % higher in homes with the highest density of agricultural use.

In this first large-scale evaluation of proximity-based metrics of agricultural glyphosate use and concentrations in house dust, we found that glyphosate was detected in 98 % of dust samples and concentrations were over 10 times higher than any of the other herbicides. Agricultural applications of glyphosate were common (67 % of homes had applications within 4 km) and were associated with up to 100 % higher concentrations after adjusting for home and garden use (Guha, 2013) and occupational exposures that were independent predictors of exposure. Concern about glyphosate exposure has increased because of the numerous animal and epidemiologic studies that have demonstrated adverse effects via multiple mechanisms (Myers, 2016). In addition, the half-life of glyphosate in soil and water is longer than previously recognized with current estimates ranging from days to several months and up to a year (Myers, 2016). Despite the important role of house dust as an exposure route for children, few studies of glyphosate exposure have been conducted and limited prior studies evaluated levels in house dust. In addition to our findings, one study of 11 homes from two rural Iowa counties showed that homes located on land used for farming had 3-times higher glyphosate concentrations in house dust relative to non-farm homes (Curwin, 2005). A study in France showed that median glyphosate concentrations were about 3-times higher in the dust of homes within 500 m of crops relative to those farther away (1410 ng/g and 457 ng/g, respectively) (Saurat, 2023). Median concentrations of glyphosate in the CCLS homes were higher than the French study (1668 ng/g in homes with agricultural glyphosate use within 0.5 km and 1196 ng/g in homes with no nearby use). Additional studies have evaluated glyphosate in urine, including a study of 71 pregnant people in Indiana that showed somewhat higher mean urinary glyphosate levels among those living in rural (4.2 ng/mL) vs. urban (3.5 mg/mL) and suburban (3.2 mg/mL) areas (Parvez, 2018) and a study of 40 pregnant people in Idaho that showed higher urinary glyphosate levels among participants who lived within a third of a mile from agricultural fields during the spray season compared to those who lived further away (0.23 μg/L vs. 0.15 μg/L) (Curl, 2023).

Though few prior exposure studies have evaluated glyphosate in dust, residential proximity to agricultural fields has been positively associated with other herbicide and fungicide detection rates or concentrations in biomarker, house dust, or wristband samples (Dereumeaux et al., 2020; Harnly, 2009; Curwin, 2005; Harley, 2019; Sammartano, 2020; Ward, 2006; Harnly, et al., 2009). A number of these studies demonstrate findings generally consistent with ours, including a study of Latina girls living in an agricultural area of California that showed the odds of detecting dacthal in silicone wristbands were 3.1 times higher for participants living within 100 m of an agricultural field (Harley, 2019). Another study in the same area of California showed that agricultural use of the fungicide iprodione in the land sections adjacent to the home was associated with concentrations in the house dust (Harnly, et al., 2009). In a *meta*-regression of published data that used distances up to 1,125 m, Deziel et al. showed that house dust herbicide and fungicide concentrations were associated with agricultural use and concentrations decreased non-linearly as the distance between the treated field and home increased (Deziel, 2017).

Only a few studies have evaluated the associations of agricultural pesticide use and concentrations measured in dust or other media at distances > 1 km (Gunier, 2014; Gunier, 2011; Harnly, et al., 2009; Gunier, 2013) and one review estimated that only 25 % of studies that used GIS-based agricultural pesticide exposure metrics explored more than one buffer size (Teysseire, 2020). Primary pesticide drift from spraying operations occurs at distances up to 1 km for aerial spraying and up to 0.8 km for ground boom applications (Chester and Ward, 1984; Frost and Ware, 1970; Byass and Lake, 1977). In our prior study of fewer pesticides and homes, we observed the strongest associations at 1.25 km but did not look at larger distances (Gunier, 2011). Here, we explored buffer sizes ranging from 0.5 to 4 km to determine the optimal distance metric for each active ingredient. Application densities were strong predictors of concentrations for most, but not all (MCPA, chlorothalonil), pesticides, and the magnitude of each association varied across the buffer sizes. Our results suggest that pesticides travel varying distances and that some may travel at least 4 km from treated fields, suggesting that for the larger distances we evaluated, secondary transport would be the most important determinant of exposure. Future exposure studies and epidemiologic studies of pesticides and disease risk should consider accounting for agricultural exposures to these pesticides at distances up to 4 km and tailoring metrics to each active ingredient. For example, percent change for glyphosate concentrations using the CPUR metric were smallest at 0.5 km, and values were larger but similar across 1 km, 2 km, and 4 km; whereas percent change for simazine was largest at 1 km and 2 km and iprodione was largest at 4 km. In contrast, percent change was strongest at 0.5 km for dacthal and dicamba with sharp declines with increasing distances.

In this study we incorporated data on agricultural land use and wind direction to improve the specificity of the CPUR metric exposure estimates. Numerous studies have evaluated the distance from homes to crop fields and the area of crop fields around the home (Nuckols, 2007; Rull and Ritz, 2003; Vannier et al., 2020) in exposure metrics for epidemiologic studies. Recognizing that meteorological conditions can impact pesticide dispersion (Pfleeger, 2006) and contribute to off-target exposures (Costanzini, et al., 2018), a few others have incorporated wind direction (Rowe, 2016; Gunier, 2018; Sagiv, 2019) into their exposure metrics. However, few studies have formally evaluated the utility of incorporating extra information into the pesticide use metric. In this study, we did not find that incorporating wind direction or information on agricultural land use resulted in substantially different findings for any of the herbicides and fungicides we evaluated, suggesting that a metric using the CPUR database is sufficient to estimate exposure for epidemiologic studies of these pesticides. Future environmental exposure studies could consider evaluation of additional pesticide active ingredients, as well as evaluation of other meteorological conditions that may influence pesticide fate and transport (i.e., wind speed, relative humidity, and air temperature) and physical properties of the pesticide active ingredients (i.e. volatility, dissipation half-life, and water solubility).

The properties of each active ingredient such as the half-life in soil, water solubility, vapor pressure, and octanol/water partition coefficient influence the ingredient’s ability to persist and be transported from agricultural land to unintended areas (Bennett et al., 1998; Farha, 2016; Gavrilescu, 2005; Juraske et al., 2008; Van Eerd et al., 2003). The herbicides that were most widely used in our study, including glyphosate, simazine, and trifluralin, have long field dissipation half-lives of over 100 days, but other active ingredients like 2,4-D, dicamba, and iprodione have shorter half-lives of less than 30 days (USDA Agricultural Research Service, 1995). Unlike our prior work that demonstrated the 60-day period between insecticide application and dust collection had the strongest predictive value (Madrigal, 2023), we did not find that varying the time period between application and dust collection in our metrics resulted in differences in prediction for the herbicides or fungicides included here. This likely indicates that these herbicides and fungicides are stable indoors with longer indoor half-lives than some of the insecticides that we evaluated previously.

To our knowledge, no prior studies have compared CPUR density metrics for ground and aerial spraying in relation to residential exposure. Although most applications of the pesticides in our study were by ground spraying methods, the volume and frequency of use of glyphosate, simazine, 2,4-D, and trifluralin allowed us to compare metrics specific to each application type for these four herbicides. For these ingredients, aerial spraying did not appear to be driving associations between the CPUR metric and dust concentration. Our findings for the separate aerial and ground spraying metrics were generally consistent with the CPUR metric that did not distinguish application method.

This study has many strengths, including the use of a comprehensive database of agricultural pesticide use with detailed records of applications of specific active ingredients in the study area. We were also able to incorporate modeled wind data for each participant residence. Using stored dust samples, we were able to measure glyphosate in dust using updated methods that relied on liquid chromatography tandem MS. The CCLS included a large number of homes, facilitating the evaluation of numerous active ingredients including those that are used infrequently. Additionally, we had detailed information from the study interview that permitted us to evaluate pesticide use in the home and garden, including on indoor plants and for lawn and garden weeds, professional lawn treatments, and pesticide exposures from adults who worked in pesticide-exposed occupations. In prior analyses in the CCLS, self-reported pesticide treatments were positively associated with levels of most active ingredients measured in the dust samples (Deziel, 2015). We were able to control for these independent predictors of dust concentration in our models, demonstrating that agricultural use of active ingredients like glyphosate, 2,4-D and trifluralin predicted dust concentrations even after controlling for home and garden use.

There are some limitations to our study. The temporal resolution of the NLCD available only included years 2001, 2004, and 2006, but our dust samples were collected over a period from 2001 to 2007. The NCLD data does not distinguish between different specific types of row crops. In California, multiple crops may be planted in a field during an annual growing period, which could result in misclassification of exposure if the annual land cover data we used did not accurately capture land use when crops changed over time or when fields had multiple crops grown in a given season or year. However, except for 365-day iprodione and chlorothalonil comparisons in the 0.5 km buffer, the correlations we observed for the CROP-A metric and CPUR metrics were high, suggesting that the land cover data are useful in the absence of the ability to conduct an exposure assessment with real-time crop mapping. When we adjusted the CPUR metric for wind direction, we used a 90-degree wind wedge that is less conservative than the 45-degree wedge prior studies have incorporated (Rowe, 2016; Gunier, 2018; Sagiv, 2019). We did not account for wind speed, which could impact primary and secondary pesticide drift; our wind dataset was at a 32 km resolution.

We did not separately evaluate metrics for applications to non-agricultural areas like rights-of-way and golf courses, which may be determinants of concentrations of pesticide active ingredients in house dust; however, the proportion of records attributed to these types of land use was small (0.1 %) relative to agricultural applications. We evaluated the contributions of aerial and ground spraying methods separately, but these data are limited in that most applications used ground spraying methods. Furthermore, the database does not provide details on the apparatus used for spraying (e.g., tractor-pulled sprayers), which for ground-spraying methods could impact the variation in the areal distribution of the off-field emissions of the active ingredient. Future exposure research could be informed by obtaining detailed information on the equipment used for spraying, but this is not currently collected in the data source we used. We acknowledge that primary and secondary pesticide drift, which occur during the active pesticide application and after it, respectively, may each contribute to pesticide residues in household dust, but we are unable to determine which of these may be responsible for the pesticide concentrations measured in the house dust. Additionally, our exposure assessment relied on pesticide concentrations measured from house dust but it is not clear how pesticide levels in house dust relate to biological levels in children and other household residents. Although we did not have biological samples from study participants that could be used in comparison, the expected half-life of pesticides in urine is short (e.g., less than 24 h for glyphosate (Connolly, 2019) whereas dust measures represent longer term exposures. Additional research is needed to evaluate the associations between dust levels and exposures measured in biological samples.

## 5. Conclusions

In this population-based study more than half of the participants had agricultural herbicide or fungicide use within 4 km of their home, largely driven by the high prevalence of glyphosate, simazine, 2,4-D, trifluralin, and iprodione applications. Our GIS-based metric created using the California pesticide use report database showed mostly high correlations with metrics adjusted for land use and wind direction and these metrics had similar associations in models to predict dust concentrations for most of the pesticides we evaluated. The observed variation in associations by buffer size indicate that GIS-based exposure metrics used in epidemiologic studies should be tailored to each active ingredient. Our findings suggest that most of these herbicides and fungicides travel from the field via primary and secondary drift to homes in the surrounding area potentially impacting the health of children and other vulnerable groups. Investigation of the health impacts of residential proximity to agricultural glyphosate use may be particularly warranted given that glyphosate was detected in almost 99 % of dust samples and concentrations were an order of magnitude higher than any other herbicide or fungicide. It should be noted that the CPUR metric is based on a robust and mandatory pesticide use reporting program that is unique to the state of California, USA. Our findings highlight the need for similar data resources in all areas where agricultural and residential land are highly integrated.

## Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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## Data availability

The study data are available upon request pending approval from the authors and relevant committees for protection of human subjects. The data are not publicly available due to privacy restrictions.

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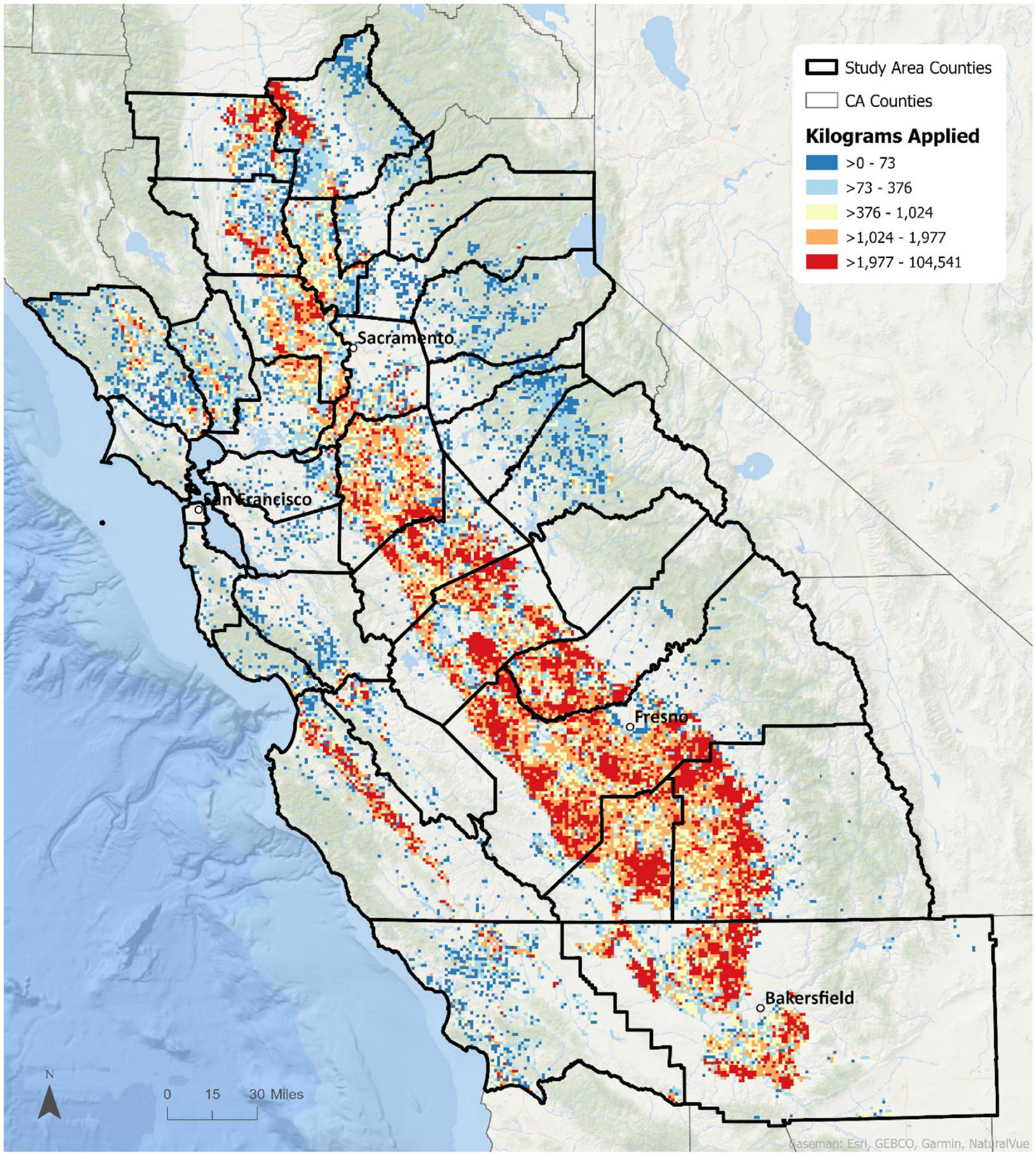

**Fig. 1.**

Distribution of agricultural herbicide and fungicide applications for 8 study herbicides and 2 study fungicides applied 2000–2006 across all study counties.

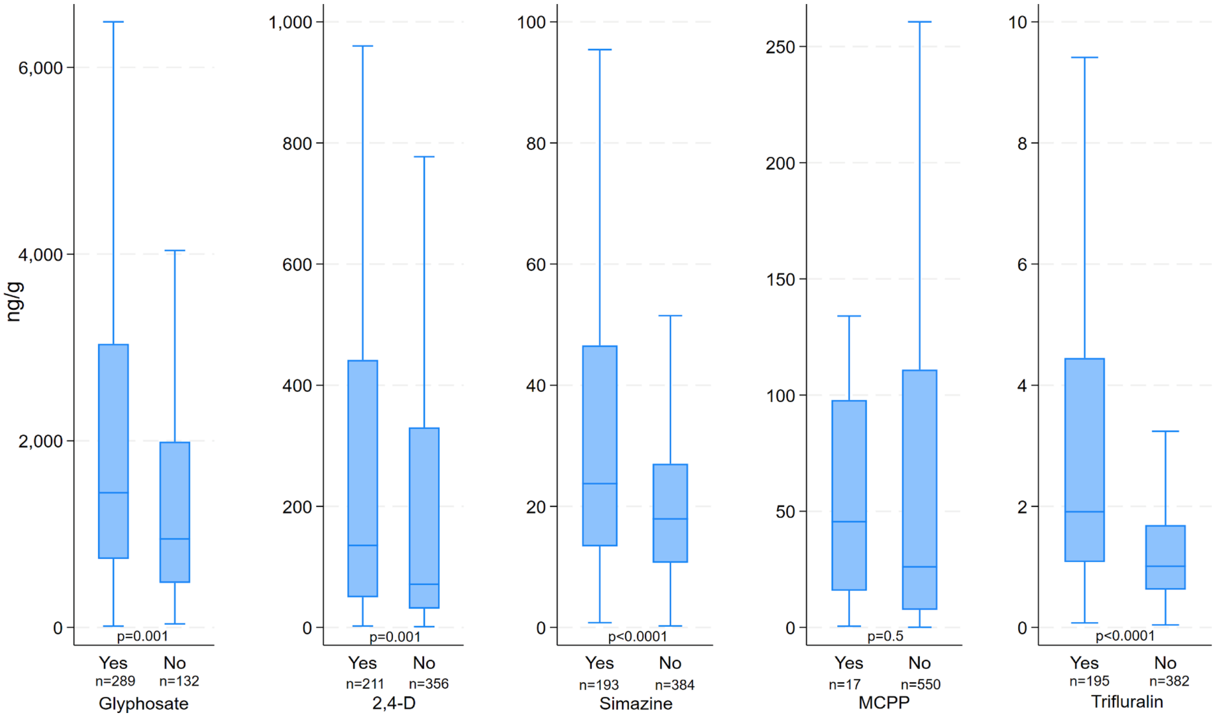

**Fig. 2.**

Distributions of concentrations in house dust of homes with (Yes) and without (No) nearby agricultural use within 4 km in the prior 365 days for five herbicides detected in ≥60 % of dust samples (glyphosate, 2,4-D, simazine, MCPP, trifluralin). 2,4-D: 2,4-dichlorophenoxyacetic acid, MCPP: 2-(4 chloro-2-methylphenoxy) propionic acid. Yes = Homes with herbicide applications (exposed); No = Homes without herbicide applications (unexposed); p-values from Wilcoxon rank sum test used to compare the distribution of the pesticide concentration in the dust among homes classified as exposed and those classified as unexposed; samples below the detection limit were assigned a value using a single imputation method. Dust concentrations with values 1.5 times the IQR above the upper quartile were excluded from the graphs.

## Table 1

Number (%) of homes with agricultural applications of herbicides and fungicides and median (25th-75th percentile) of the CPUR[a] exposure metric (density of use in kg/km$^2$) within 0.5 km and 4 km during the 365 days prior to sample collection for n = 578 homes.

| | **0.5 km buffer** | | **4 km buffer** | |
|---|---|---|---|---|
| **Herbicide** | **N (%)** | **Median (25th-75th)** | **N (%)** | **Median (25th-75th)** |
| Glyphosate | 156 (27.0) | 5.5 (0.9–19.6) | 385 (66.6) | 2.7 (0.1–14.0) |
| Simazine | 54 (9.3) | 3.1 (0.7–8.3) | 193 (33.4) | 1.3 (0.2–5.1) |
| 2,4-D | 51 (8.8) | 2.8 (0.9–6.5) | 215 (37.2) | 0.9 (0.1–3.1) |
| Trifluralin | 38 (6.6) | 4.8 (1.9–10.3) | 195 (33.7) | 0.8 (0.1–3.6) |
| MCPA | 28 (4.8) | 3.4 (0.6–7.9) | 154 (26.6) | 0.6 (0.2–1.7) |
| Dacthal | 14 (2.4) | 12.2 (1.5–27.5) | 53 (9.2) | 1.9 (0.2–8.7) |
| Dicamba | 12 (2.1) | 0.5 (0.1–3.2) | 83 (14.4) | 0.1 (0.03–0.3) |
| MCPP | 1 (<1) | 0.1[b] | 17 (2.9) | 0.1 (0.01–0.2) |
| **Fungicide** | | | | |
| Iprodione | 71 (12.3) | 1.4 (0.3–4.9) | 236 (40.8) | 0.5 (0.04–2.2) |
| Chlorothalonil | 39 (6.8) | 2.7 (0.4–6.3) | 196 (34.0) | 0.7 (0.1–3.3) |

2,4-D: 2,4-dichlorophenoxyacetic acid.

MCPA: 2-methyl-4-chlorophenoxyacetic acid.

MCPP: 2-(4 chloro-2-methylphenoxy) propionic acid.

[a] CPUR = CPUR Metric (kg/km$^2$): herbicide or fungicide use density proportional to the area of the buffer.

[b] Value for 1 home with agricultural use of MCPP within the buffer.

## Table 2

Detection limits, percent detections, and concentrations for eight herbicides and two fungicides measured in dust samples, overall and by agricultural use of the active ingredient within 4 km of the home in the 365 days prior to dust collection.

| Herbicide | Detection limit (ng/g dust) | Detected, n (%) | Median (IQR) concentration (ng/g dust)[d] | Agricultural Use ≤ 4 km | | No Agricultural Use ≤ 4 km | |
|---|---|---|---|---|---|---|---|
| | | | | Detected, n (%) | Median (IQR) concentration (ng/g dust)[d] | Detected, n (%) | Median (IQR) concentration (ng/g dust)[d] |
| Glyphosate[a] | 47 | 415 (98.6) | 1277 (670–2775) | 285 (98.6) | 1495 (754–3043) | 130 (98.5) | 966 (482–2008) |
| 2,4-D[b] | 5 | 553 (97.7) | 103.9 (36.8–426.4) | 209 (99.0) | 135.7 (52.3–441.5) | 344 (96.9) | 82.9 (32.6–366.7) |
| Simazine[c] | 2 | 522 (90.5) | 20.6 (13.9–36.4) | 180 (93.3) | 25.5 (15.5–48.1) | 342 (89.1) | 19.5 (13.2–28.9) |
| MCPP[b] | 5 | 478 (84.3) | 39.5 (14.9–135.2) | 16 (94.1) | 57.5 (16.8–102.8) | 462 (84.0) | 38.6 (14.5–139.0) |
| Trifluralin[c] | 2 | 353 (61.2) | 1.8 (1.0–4.2) | 159 (81.5) | 2.4 (1.4–5.2) | 194 (50.8) | 1.4 (0.9–2.8) |
| Dacthal[c] | 1 | 204 (35.4) | 2.5 (1.5–4.9) | 39 (76.6) | 5.1 (2.8–12.5) | 165 (31.5) | 2.1 (1.4–3.8) |
| Dicamba[b] | 5 | 158 (27.9) | 7.7 (4.4–16.8) | 31 (38.3) | 6.7 (3.0–22.9) | 127 (26.1) | 7.7 (4.5–16.5) |
| MCPA[b] | 5 | 88 (15.5) | 19.6 (12.6–34.7) | 32 (21.3) | 19.1 (11.7–33.8) | 56 (13.4) | 21.1 (13.1–35.4) |
| **Fungicide** | | | | | | | |
| Chlorothalonil[c] | 10 | 108 (18.8) | 21.8 (11.8–47.5) | 38 (19.4) | 19.9 (12.1–54.4) | 70 (18.4) | 23.6 (10.7–47.2) |
| Iprodione[c] | 20 | 70 (12.1) | 48.5 (28.6–91.2) | 54 (22.9) | 47.8 (25.4–78.7) | 16 (4.7) | 54.2 (37.7–117.1) |

IQR: interquartile range.

MCPA: 2-methyl-4-chlorophenoxyacetic acid.

MCPP: 2-(4 chloro-2-methylphenoxy) propionic acid.

2,4-D: 2,4-dichlorophenoxyacetic acid.

[a] n = 421 homes with dust measurements using formic acid in methanol method; 24 % measured from the household vacuum.

[b] n = 567 homes with dust measurements using acid herbicide method; 34 % measured from the household vacuum.

[c] n = 577 homes with dust measurements using hexane:acetone method; 29 % measured from the household vacuum.

[d] Distribution among residences with pesticide concentrations above the detection limit.

## Table 3

Multivariable associations[a] of the 365-day CPUR, CROP-A, and W-CPUR metrics with concentrations of glyphosate, simazine, 2,4-D, and trifluralin.

| Herbicide | Buffer | CPUR[b] |  | CROP-A[c] |  | W-CPUR[d] |  |
|---|---|---|---|---|---|---|---|
|  |  | Density (kg/km$^2$) vs. 0 | % Increase (95 % CI) | Density (kg/km$^2$) vs. 0 | % Increase (95 % CI) | Density (kg/km$^2$) vs. 0 | % Increase (95 % CI) |
| **Glyphosate** [e] | ***0.5 km*** | >0-≤ 2.2 | −12 (−39, 27) | >0-≤ 2.0 | 40 (−9, 114) | >0-≤ 0.1 | − 6 (−35, 36) |
|  |  | 2.2–11.7 | 88 (31, 171) | 2.0–17.0 | 137 (53, 268) | 0.1–2.5 | 46 (1, 113) |
|  |  | > 11.7–207 | 83 (26, 167) | > 17.0–210 | 66 (7, 155) | > 2.5–143 | 116 (49, 214) |
|  |  | p-trend | 0.0005 | p-trend | 0.02 | p-trend | <0.0001 |
|  | ***1 km*** | >0-≤ 1.3 | 13 (−18, 56) | >0-≤ 1.3 | 45 (3, 106) | >0-≤ 0.2 | 9 (−21, 51) |
|  |  | 1.3–9.9 | 49 (7, 107) | 1.3–10.7 | 86 (31, 164) | 0.2–1.7 | 72 (23, 142) |
|  |  | > 9.9–187 | 109 (50, 192) | > 10.7–187 | 126 (58, 222) | > 1.7–74 | 95 (40, 172) |
|  |  | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | 0.0001 |
|  | ***2 km*** | >0-≤ 1.4 | − 5 (−30, 28) | >0-≤ 2.6 | 29 (−5, 76) | >0-≤ 0.2 | −10 (−34, 21) |
|  |  | 1.4–10.1 | 51 (12, 104) | 2.6–12.4 | 60 (16, 119) | 0.2–2.1 | 74 (28, 135) |
|  |  | > 10.1–113 | 111 (55, 188) | > 12.4 –113 | 149 (80, 244) | > 2.1–56 | 98 (45, 168) |
|  |  | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |
|  | ***4 km*** | >0-≤ 0.4 | − 9 (−32, 21) | >0-≤ 1.6 | 22 (−9, 64) | >0-≤ 0.1 | −1 (−26, 33) |
|  |  | 0.4–9.2 | 25 (−7, 68) | 1.6–13.4 | 43 (6, 91) | 0.1–2.0 | 34 (0, 80) |
|  |  | > 9.2–699 | 110 (55, 183) | > 13.4–700 | 144 (81, 229) | > 2.0–124 | 102 (50, 171) |
|  |  | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |
|  | ***0.5 km*** | >0-≤3.1 | 168 (62, 345) | >0-≤2.8 | 210 (80, 435) | >0-≤0.6 | 108 (25, 246) |
|  |  | >3.1–70 | 163 (58, 337) | >2.8–72 | 153 (44, 345) | >0.6–31 | 230 (98, 452) |
|  |  | p-trend | 0.0002 | p-trend | 0.001 | p-trend | <0.0001 |
|  | ***1 km*** | >0-≤2.1 | 82 (20, 174) | >0-≤2.1 | 148 (61, 282) | >0-≤0.4 | 69 (12, 156) |
|  |  | >2.1–60 | 239 (124, 415) | >2.1–58 | 197 (92, 359) | >0.4–23 | 278 (149, 475) |
|  |  | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |
| **Simazine** [f] | ***2 km*** | >0-≤ 0.5 | 21 (−19, 82) | >0-≤ 0.5 | 43 (−5, 117) | >0-≤ 0.1 | 28 (−14, 93) |
|  |  | 0.5–3.2 | 72 (15, 157) | 0.5–3.5 | 82 (20, 175) | 0.1–0.7 | 70 (13, 155) |
|  |  | > 3.2–76 | 241 (129, 408) | > 3.5–75 | 237 (123, 408) | > 0.7–27 | 237 (125, 405) |
|  |  | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |

| Herbicide | Buffer | CPUR[b] | | CROP-A[c] | | W-CPUR[d] | |
|---|---|---|---|---|---|---|---|
| | | Density (kg/km$^2$) vs. 0 | % Increase (95 % CI) | Density (kg/km$^2$) vs. 0 | % Increase (95 % CI) | Density (kg/km$^2$) vs. 0 | % Increase (95 % CI) |
| | ***4 km*** | >0-≤ 0.4 | 48 (5, 109) | >0-≤ 0.5 | 44 (1, 105) | >0-≤ 0.1 | 51 (7, 114) |
| | | 0.4–3.2 | 23 (−13, 74) | 0.5–3.6 | 23 (−14, 76) | 0.1–0.8 | 30 (−8, 84) |
| | | > 3.2–91 | 199 (112, 323) | > 3.6–92 | 213 (120, 347) | > 0.8–21 | 209 (118, 337) |
| | | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |
| | ***0.5 km*** | >0 | 60 (0, 156) | >0 | 68 (0, 181) | >0 | 33 (−18, 117) |
| | ***1 km*** | >0 | 83 (26, 166) | >0 | 75 (18, 158) | >0 | 72 (18, 149) |
| | ***1 km*** | >0-≤1.4 | 60 (−2, 163) | >0-≤1.5 | 59 (−6, 170) | >0-≤0.2 | 39 (−16, 131) |
| | | >1.4–46 | 112 (27, 253) | >1.5–44 | 92 (12, 230) | >0.2–8.9 | 111 (27, 251) |
| | | p-trend | 0.005 | p-trend | 0.002 | p-trend | 0.003 |
| | ***2 km*** | >0 | 42 (4, 94) | | 35 (−2, 85) | | 45 (6, 98) |
| 2,4-D[g] | ***2 km*** | >0-≤1.0 | −3 (−36, 45) | >0-≤1.1 | −16 (−44, 28) | >0-≤0.2 | 19 (−21, 79) |
| | | >1.0–31 | 108 (38, 212) | >1.1–31 | 117 (43, 230) | >0.2–8.8 | 78 (18, 169) |
| | | p-trend | 0.0003 | p-trend | 0.0003 | p-trend | 0.01 |
| | ***4 km*** | >0 | 47 (12, 94) | >0 | 41 (7, 87) | >0 | 43 (9, 88) |
| | ***4 km*** | >0-≤ 0.3 | 5 (−30, 57) | >0-≤ 0.4 | 6 (−31, 61) | >0-≤ 0.02 | 20 (−20, 81) |
| | | 0.3–2.2 | 54 (2, 131) | 0.4–2.3 | 57 (2, 140) | 0.02–0.5 | 22 (−19, 84) |
| | | > 2.2–16 | 102 (33, 206) | > 2.3–16 | 71 (12, 160) | > 0.5–4.6 | 98 (32, 198) |
| | | p-trend | 0.001 | p-trend | 0.003 | p-trend | 0.01 |
| | ***0.5 km*** | >0-≤4.8 | 224 (63, 544) | >0-≤5.3 | 217 (42, 604) | >0-≤0.4 | 351 (127, 797) |
| | | >4.8–98 | 578 (242, 1244) | >5.3–98 | 628 (217, 1569) | >0.4–22 | 408 (151, 928) |
| | | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |
| **Trifluralin**[h] | ***1 km*** | >0-≤2.3 | 202 (80, 406) | >0-≤1.4 | 309 (135, 612) | >0-≤0.3 | 163 (55, 344) |
| | | >2.3–86 | 486 (249, 886) | >1.4–89 | 446 (212, 856) | >0.3–19 | 559 (290, 1015) |
| | | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |
| | ***2 km*** | >0-≤1.1 | 121 (47, 231) | >0-≤1.4 | 118 (42, 235) | >0-≤0.2 | 135 (56, 254) |
| | | >1.1–55 | 478 (288, 760) | >1.4–61 | 487 (286, 791) | >0.2–18 | 435 (258, 701) |
| | | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |
| | ***4 km*** | >0-≤ 0.2 | 44 (−3, 114) | >0-≤ 0.3 | 67 (11, 150) | >0-≤ 0.02 | 58 (6, 135) |
| | | 0.2–2.2 | 151 (70, 272) | 0.3–2.7 | 133 (55, 248) | 0.02–0.4 | 131 (56, 242) |
| | | > 2.2–41 | 524 (328, 810) | > 2.7–37 | 560 (347, 873) | > 0.4–11 | 532 (332, 825) |

| Herbicide | Buffer | CPUR[b] | | CROP-A[c] | | W-CPUR[d] | |
|---|---|---|---|---|---|---|---|
| | | **Density ($kg/km^2$) vs. 0** | **% Increase (95 % CI)** | **Density ($kg/km^2$) vs. 0** | **% Increase (95 % CI)** | **Density ($kg/km^2$) vs. 0** | **% Increase (95 % CI)** |
| | | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |

[a] Individual Tobit regression models were used to model the natural log transformed dust concentrations with a forward approach to evaluate covariates. Any covariate with a p-value ≤ 0.1 was retained in the final multivariable model. We reported 100 %*(exp(β)-1) to show the percent increase in the dust sample concentration comparing categories of the CPUR, CROP-A, and W-CPUR metrics to no use (i.e., a reference group of 0 $kg/km^2$ of agricultural use according to the database).

[b] CPUR = CPUR Metric ($kg/km^2$): herbicide use density proportional to the area of the buffer.

[c] CROP-A = Crop area adjusted CPUR Metric ($kg/km^2$): herbicide use density proportional to the area of cultivated crops and pasture/hay with herbicide use in the buffer.

[d] W-CPUR = Wind-adjusted CPUR Metric ($kg/km^2$): herbicide use density proportional to the area of the buffer, weighted to account for the proportion of days the residence was downwind of the section in which the herbicide was applied during the period between application and dust collection

[e] Adjusted for variables associated (positively) with glyphosate concentrations: season, treatments for bees and lawn/garden weeds, and occupations involving farming and gardening.

[f] Adjusted for treatments for flies and year; all were positively associated with simazine concentrations.

[g] Adjusted for the use of treatments for lawn/garden weeds that were positively associated with 2,4-D concentrations. Farm worker occupation was inversely associated.

[h] Adjusted for shoe removal that was inversely associated with concentrations and treatments for lawn/garden weeds that was positively associated with trifluralin concentrations.

## Table 4

Multivariable associations[a] of the 365-day CPUR, CROP-A, and W-CPUR metrics with odds of MCPA, dacthal, dicamba, iprodione, and chlorothalonil detections in house dust.

| Herbicide or fungicide | Buffer | CPUR[b] | | CROP-A[c] | | W-CPUR[d] | |
|---|---|---|---|---|---|---|---|
| | | **Density (kg/km$^2$) vs. 0** | **OR (95 % CI) detect vs. non-detect** | **Density (kg/km$^2$) vs. 0** | **OR (95 % CI) detect vs. non-detect** | **Density (kg/km$^2$) vs. 0** | **OR (95 % CI) detect vs. non-detect** |
| **MCPA** [e] | ***0.5 km*** | >0 | 2.1 (0.8–5.5) | >0 | 2.2 (0.8–5.8) | >0 | 2.2 (0.8–5.8) |
| | ***1 km*** | >0 | 1.9 (0.9–4.1) | >0 | 2.2 (1.0–4.9) | >0 | 2.0 (0.9–4.3) |
| | ***2 km*** | >0 | 2.0 (1.1–3.5) | >0 | 2.1 (1.2–3.7) | >0 | 2.0 (1.1–3.5) |
| | | >0-≤0.7 | 2.5 (1.2–5.1) | >0-≤0.7 | 3.0 (1.5–6.1) | >0-≤0.1 | 2.1 (1.0–4.5) |
| | | >0.7–10.4 | 1.5 (0.6–3.4) | >0.7–10.3 | 1.2 (0.5–3.1) | >0.1–4.0 | 1.8 (0.8–4.1) |
| | | p-trend | 0.41 | p-trend | 0.65 | p-trend | 0.18 |
| | ***4 km*** | >0 | 1.9 (1.1–3.1) | >0 | 2.0 (1.2–3.3) | >0 | 1.9 (1.2–3.2) |
| | | >0-≤ 0.3 | 1.6 (0.8–3.4) | >0-≤ 0.3 | 2.2 (1.1–4.5) | >0-≤ 0.04 | 1.9 (0.9–3.9) |
| | | 0.3–1.1 | 2.3 (1.1–4.8) | 0.3–1.2 | 1.8 (0.8–4.1) | 0.04–0.3 | 1.6 (0.7–3.6) |
| | | > 1.1–8.9 | 1.9 (0.9–4.1) | > 1.2–8.9 | 2.0 (0.9–4.4) | > 0.3–3.7 | 2.4 (1.1–5.1) |
| | | p-trend | 0.07 | p-trend | 0.07 | p-trend | 0.04 |
| **Dacthal** [f] | ***0.5 km*** | >0 | 24.7 (3.2–190.7) | >0 | –[j] | >0 | 22.5 (2.9–175.3) |
| | ***1 km*** | >0 | 9.6 (3.2–28.6) | >0 | –[j] | >0 | 9.6 (3.2–28.6) |
| | ***2 km*** | >0 | 9.5 (3.6–25.5) | >0 | 11.3 (3.8–33.4) | >0 | 9.5 (3.6–25.5) |
| | ***4 km*** | >0 | 5.9 (3.1–11.2) | >0 | 6.6 (3.3–13.4) | >0 | 5.7 (3.0–10.9) |
| **Dicamba** [g] | ***0.5 km*** | >0 | 4.2 (1.3–13.7) | >0 | 3.9 (1.0–15.2) | >0 | 4.2 (1.3–13.7) |
| | ***1 km*** | >0 | 2.3 (0.9–5.5) | >0 | 2.5 (0.9–6.7) | >0 | 2.1 (0.8–5.1) |
| | ***2 km*** | >0 | 3.0 (1.6–5.8) | >0 | 2.9 (1.5–5.8) | >0 | 2.8 (1.4–5.6) |
| | ***4 km*** | >0 | 1.9 (1.1–3.1) | >0 | 2.0 (1.2–3.4) | >0 | 1.9 (1.1–3.2) |
| | | >0-≤0.1 | 2.2 (1.1–4.2) | >0-≤0.1 | 2.3 (1.2–4.7) | >0-≤0.02 | 2.8 (1.4–5.6) |
| | | >0.1–5.3 | 1.6 (0.8–3.2) | >0.1–5.3 | 1.8 (0.9–3.7) | >0.02–1.8 | 1.2 (0.6–2.6) |
| | | p-trend | 0.17 | p-trend | 0.10 | p-trend | 0.72 |
| **Iprodione** [h] | ***0.5 km*** | >0-≤1.4 | 4.9 (2.1–11.5) | >0-≤1.0 | 4.6 (1.9–11.2) | >0-≤0.2 | 5.2 (2.2–12.0) |
| | | >1.4–51.5 | 5.5 (2.4–12.3) | >1.0–18.6 | 6.5 (2.7–15.9) | >0.2–14.2 | 5.8 (2.6–13.2) |

| Herbicide or fungicide | Buffer | CPUR[b] | | CROP-A[c] | | W-CPUR[d] | |
|---|---|---|---|---|---|---|---|
| | | **Density (kg/km$^2$) vs. 0** | **OR (95 % CI) detect vs. non-detect** | **Density (kg/km$^2$) vs. 0** | **OR (95 % CI) detect vs. non-detect** | **Density (kg/km$^2$) vs. 0** | **OR (95 % CI) detect vs. non-detect** |
| | | p-trend | 0.0001 | p-trend | <0.0001 | p-trend | 0.0001 |
| | ***1 km*** | >0-≤1.1 | 6.1 (2.9–12.6) | >0-≤1.0 | 7.4 (3.5–15.9) | >0-≤0.1 | 6.7 (3.2–13.9) |
| | | >1.1–31.7 | 5.4 (2.6–11.2) | >1.0–32.4 | 6.6 (3.1–14.0) | >0.1–15.4 | 5.4 (2.6–11.2) |
| | | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |
| | ***2 km*** | >0-≤ 0.2 | 3.3 (1.4–7.9) | >0-≤ 0.4 | 4.6 (2.0–10.9) | >0-≤ 0.03 | 3.1 (1.3–7.5) |
| | | 0.2–1.9 | 5.9 (2.7–12.7) | 0.4–2.2 | 9.3 (4.3–20.3) | 0.03–0.3 | 6.8 (3.2–14.7) |
| | | > 1.9–44.3 | 7.8 (3.7–16.4) | > 2.2–44.8 | 6.2 (2.8–13.8) | > 0.3–20.0 | 7.5 (3.6–16.0) |
| | | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |
| | ***4 km*** | >0-≤ 0.1 | 3.4 (1.5–7.8) | >0-≤ 0.2 | 1.9 (0.8–4.7) | >0-≤ 0.01 | 2.3 (1.0–5.7) |
| | | 0.1–1.4 | 3.3 (1.5–7.5) | 0.2–1.7 | 4.8 (2.3–10.2) | 0.01–0.3 | 4.5 (2.1–10.0) |
| | | > 1.4–30.7 | 9.8 (4.7–20.1) | > 1.7–29.9 | 8.0 (4.0–16.2) | > 0.3–13.3 | 10.3 (5.0–21.1) |
| | | p-trend | <0.0001 | p-trend | <0.0001 | p-trend | <0.0001 |
| **Chlorothalonil** [i] | ***0.5 km*** | >0 | 0.8 (0.3–1.9) | >0 | 0.5 (0.1–1.7) | >0 | 0.8 (0.3–2.0) |
| | ***1 km*** | >0 | 0.9 (0.5–1.8) | >0 | 1.0 (0.5–2.1) | >0 | 1.0 (0.5–1.9) |
| | | >0-≤1.2 | 0.8 (0.3–2.1) | >0-≤1.3 | 1.1 (0.4–3.0) | >0-≤0.2 | 0.8 (0.3–2.2) |
| | | >1.2–25.4 | 1.1 (0.4–2.5) | >1.3–50.2 | 0.9 (0.3–2.5) | >0.2–9.3 | 1.1 (0.5–2.6) |
| | | p-trend | 0.88 | p-trend | 0.84 | p-trend | 0.81 |
| | ***2 km*** | >0 | 1.1 (0.7–1.9) | >0 | 1.2 (0.7–2.1) | >0 | 1.1 (0.7–1.9) |
| | | >0-≤1.2 | 0.9 (0.5–1.9) | >0-≤1.6 | 1.1 (0.5–2.3) | >0-≤0.1 | 1.1 (0.5–2.1) |
| | | >1.2–27.3 | 1.3 (0.7–2.5) | >1.6–20.3 | 1.3 (0.6–2.6) | >0.1–18.3 | 1.2 (0.6–2.3) |
| | | p-trend | 0.42 | p-trend | 0.47 | p-trend | 0.65 |
| | ***4 km*** | >0 | 1.0 (0.6–1.6) | >0 | 0.9 (0.6–1.5) | >0 | 1.0 (0.6–1.6) |
| | | >0-≤ 0.2 | 1.0 (0.5–2.0) | >0-≤ 0.4 | 1.0 (0.5–2.0) | >0-≤ 0.02 | 1.1 (0.6–2.2) |
| | | 0.2–1.9 | 0.9 (0.4–1.8) | 0.4–2.7 | 0.9 (0.4–1.9) | 0.02–0.5 | 0.8 (0.4–1.6) |
| | | > 1.9–23.5 | 1.1 (0.6–2.1) | > 2.7–23.3 | 0.9 (0.5–1.9) | > 0.5–15.9 | 1.1 (0.6–2.2) |
| | | p-trend | 0.83 | p-trend | 0.89 | p-trend | 0.73 |

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[a] Individual logistic regression models were used to model the odds of detection with a forward approach to evaluate covariates. Any covariate with a p-value ≤ 0.1 was retained in the final multivariable model. All comparisons use a reference group of 0 kg/km$^2$ of agricultural use according to the database.

[b] CPUR = CPUR Metric (kg/km$^2$): herbicide or fungicide use density proportional to the area of the buffer.

[c] CROP-A = Crop area adjusted CPUR Metric ($kg/km^2$): herbicide or fungicide use density proportional to the area of cultivated crops and pasture/hay with herbicide or fungicide use in the buffer.

[d] W-CPUR = Wind-adjusted CPUR Metric ($kg/km^2$): herbicide or fungicide use density proportional to the area of the buffer, weighted to account for the proportion of days the residence was downwind of the section in which the herbicide or fungicide was applied during the period between application and dust collection

[e] Adjusted for professional outdoor lawn treatments, year, and season which were positively associated with MCPA concentrations.

[f] Adjusted for agricultural packing occupations that were positively associated with dacthal concentrations.

[g] Adjusted for indoor professional pest treatments and the use of treatments for lawn and garden weeds that were positively associated with dicamba concentrations.

[h] Adjusted for season, professional outdoor lawn treatments, and farm worker occupations; all were positively associated with iprodione concentrations.

[i] Adjusted for the use of treatments for indoor plants that was positively associated and year that was inversely associated with chlorothalonil concentrations.

[j] Dashed line (–) indicates that model did not converge; 0.5 km dacthal CROP-A n = 8 homes with agricultural use and dust detections; 1 km dacthal CROP-A n = 16 homes with agricultural use and dust detections.