Agricultural Pesticide Use in California: Pesticide Prioritization, Use Densities, and Population Distributions for a Childhood Cancer Study

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Several studies have suggested an association between childhood cancer and pesticide exposure. California leads the nation in agricultural pesticide use. A mandatory reporting system for all agricultural pesticide use in the state provides information on the active ingredient, amount used, and location. We calculated pesticide use density to quantify agricultural pesticide use in California block groups for a childhood cancer study. Pesticides with similar toxicologic properties (probable carcinogens, possible carcinogens, genotoxic compounds, and developmental or reproductive toxicants) were grouped together for this analysis. To prioritize pesticides, we weighted pesticide use by the carcinogenic and exposure potential of each compound. The top-ranking individual pesticides were propargite, methyl bromide, and trifuralin. We used a geographic information system to calculate pesticide use density in pounds per square mile of total land area for all United States census-block groups in the state. Most block groups (77%) averaged less than 1 pound per square mile of use for 1991–1994 for pesticides classified as probable human carcinogens. However, at the high end of use density (> 90th percentile), there were 493 block groups with more than 569 pounds per square mile. Approximately 170,000 children under 15 years of age were living in these block groups in 1990. The distribution of agricultural pesticide use and number of potentially exposed children suggests that pesticide use density would be of value for a study of childhood cancer. Key words: agriculture, childhood cancer, ecologic study, epidemiologic study, exposure assessment, geographic information systems, pesticides, risk assessment. Environ Health Perspect 109:1071–1078 (2001). [Online 1 October 2001] http://ehpnet1.niehs.nih.gov/docs/2001/109p1071-1078gunier/abstract.html

Some epidemiologic studies suggest an association between pesticide exposure and childhood cancer (1,2). Most studies have used questionnaires to evaluate parental occupational exposure around the time of the child’s birth and exposure to the parents or child from pesticide use in the home and garden. Such information is potentially limited by response bias. Childhood cancer has not been evaluated with respect to potential exposure to agricultural pesticides because respondents are unlikely to have specific knowledge about pesticide use on nearby fields.

Few tools exist for identifying regions with a high density of agricultural pesticide use. County crop acreage totals are available, but land use varies tremendously within California counties because of differences in topography and urbanization. Some investigators have used satellite imagery and a geographic information system (GIS) to identify the location of agricultural fields (3,4). These indices provide information on the population living near fields, but only indirect estimates of pesticide use based on crop type. The resulting pesticide use estimates are limited by crop misclassification and the assumption that all fields are treated similarly for a given crop. Some studies of cancer in adults have been conducted with pesticide use data summarized at the county level (5–7). However, the number of residents living near agricultural fields and the amount of specific pesticides applied agriculturally in these communities have generally not been available.

In 1992, California accounted for 22% of all agricultural pesticide use in the United States (8). There has been some form of pesticide use reporting in California for several decades, although before 1990 reporting was limited to applications that were restricted and required a permit. The California legislature mandated the Pesticide Use Report (PUR) system in 1990 (9), legally requiring growers and applicators to report all commercial agricultural pesticide use. Every month, written or electronic records of all pesticide applications are submitted to the county agricultural commissioners. The California Department of Pesticide Regulation (Sacramento) collects the data entered by the counties and after checking for errors makes it available to the public annually for a small fee. Few states have a full pesticide use reporting system and no other state has been required to report all commercial agricultural use. Inert ingredients, which might also be toxic, are not reported. The active ingredients, which we refer to as pesticides, range from compounds with no known carcinogenic potential to substances known to cause cancer in laboratory animals (10).

The PUR data provide an opportunity to develop more geographically precise estimates of agricultural pesticide use, which may be evaluated in conjunction with cancer incidence rates. California is particularly suited for such an analysis because it also has a statewide cancer reporting system. We focused on potential exposures to children because the latency period for childhood cancer is shorter than for adult cancer. For this statewide analysis, we grouped pesticides into toxicologic categories and chemical classes to account for compounds that might act similarly in the human body or in the environment. In addition, we prioritized individual pesticides by weighting the reported pounds of use by the potential of the pesticide to cause cancer and the possibility of exposure based on volatility and environmental persistence. The geographic boundaries for which agricultural pesticide use is reported in California do not match the census boundaries. We developed GIS methods to summarize agricultural pesticide use by census-block group and estimated the number of children living in the upper 10th percentile of pesticide use density. Although we focused on childhood cancer and potential carcinogens, these methods could be modified for other health outcomes and populations.

Methods

PUR data. We used the 1991–1994 PUR data to coincide with the time period of the census and cancer incidence data, and because it represents the first few years of full pesticide use reporting. The PUR database provides the active ingredient, quantity applied, acres treated, crop treated, and date and location...
for all agricultural pesticide applications. The locations of pesticide applications are reported using an identifier that represents a section within the Public Land Survey System (PLSS). The PLSS is a nationwide survey that grids the land in each state into approximately 1-square-mile rectangular units called sections. Some areas of California were not surveyed when California became a state because of Spanish land grants. We used a version of the PLSS with the grid lines extended to cover any areas that were not surveyed (11). We checked for and deleted from further analysis applications with reported section identifiers that did not correspond to a valid section identifier within the PLSS.

A small percentage of data entry errors have been reported in the PUR that result in erroneously large amounts of pounds applied (12). We developed methods to identify and correct errors in the quantity of pesticide applied that could misclassify exposure. We used the application rate (pounds per acre) to identify potential reporting errors with unreasonably high quantities of pesticide applied. We calculated the mean application rate for each pesticide using the 1995 PUR data. We used the 1995 data for quality control because this was the most recent year available and had the fewest number of extremely high application rates. We assigned each pesticide an estimated maximum allowable application rate that was at least 2 standard deviations above its 1995 mean rate. Application rates above the estimated maximum allowable are generally so large as to be economically unfeasible. An application in the PUR database was considered an error if the application rate was greater than the maximum allowable rate for that pesticide. We checked these errors in two counties and found that they were largely the result of entry errors or illegible reporting from the growers (13). We recalculated the quantity of pesticide applied for these potentially erroneous applications by multiplying the acres treated by the maximum allowable rate.

Pesticide use by groups. We combined pesticides from the PUR data into four toxicologic groups for our statewide analysis: probable carcinogens, possible carcinogens, genotoxic compounds, and reproductive or developmental toxicants. We identified 73 pesticides for these four groups from all active ingredients reported to the PUR statewide from 1991 to 1994 (Table 1). Some individual pesticides were included in more than one group. The U.S. Environmental Protection Agency (EPA) classifies most pesticides according to their human carcinogenic potential (10). California banned or severely restricted the agricultural use of all pesticides classified as known human carcinogens (class A) or probable human carcinogens with limited human evidence (class B1) before 1991. For the purposes of this study, we created one toxicologic group with 19 pesticides classified as probable human carcinogens with sufficient evidence in laboratory animals (class B2). We formed a second group with 37 compounds categorized as possible human carcinogens with limited evidence in animals (class C).

Some pesticides are not classified as carcinogens but have evidence of other types of toxicity that may be relevant. Genotoxic chemicals have demonstrated the ability to directly damage DNA. Several laboratory tests are commonly used to assess genetic toxicity, including gene mutation, chromosome aberration, sister chromatid exchange, and DNA damage. We chose 27 pesticides with at least two positive results in genetic toxicity assays for a third toxicologic group (14,15). Because many childhood cancer cases occur shortly after birth and may be related to perinatal exposures, reproductive and developmental toxicants were also of interest. We selected 19 pesticides with at least one positive result in reproductive or developmental studies conducted in laboratory animals to form a final group for analysis (16).

We combined pesticides into four additional groups based on chemical class (organochlorides, organophosphates, carbamates, and dithiocarbamates). We identified chemical classes using a pesticide dictionary and chemical structure (17). There were 36 pesticides with reported use between 1991 and 1994 in these four classes. Organochloride insecticides had mostly been replaced by organophosphates by 1990, so these represent the smallest and largest groups, respectively. Table 1 provides a list of pesticides in each chemical class.

Table 1. Pesticides with reported use in California, 1991–1994, in toxicologic and chemical groups.

| Toxicologic and chemical groups | Pesticides |
|---------------------------------|------------|
| Probable carcinogens (class B2)² | Alachlor, cacodylic acid, captan, chlordane, chlorothalonil, dinomadize, 1,3-dichloropropene, iprodione, lindane, mancozeb, maneb, metat sodium, orthophenylphenol, oxythioxinax, propargite, propoxur, benthalchlorophenol, propyzamide, vinclozolin |
| Possible carcinogens (class C)² | Acephate, acetol, amitraz, atrazine, benomyl, bifenthrin, bromacil, bromoxynil, carbyl, chlor-thal-dimethyl, cyazine, cypermethrin, dichlofenil, dichlorvos, dicloflop-methyl, dicrofen, dimethoate, ethallinfluor, fosetyl-al, hydrogen cyanamide, imazalil, isophor, methidathion, metolachlor, molinate, norfuralon, oryzalin, oxadiazon, oxyfluorfen, pendimethalin, permethrin, phosmet, phoshamidon, piperonyl butoxide, simazine, triadimefon, trifluralin |
| Genotoxic compounds² | 2,4-Diethylamidine, acephate, alachlor, aldicarb, atrazine, benomyl, captan, carbyl, carbafuran, chlordane, chloropicrin, chlorothalonil, chloropyrifos, diazinon, 1,3-dichloropropene, diquat dibromide, malation, metat sodium, methyl bromide, methyl parathion, mevinphos, orthophenylphenol, oxydemeton, methyl, parquat dichloride, pentachlorophenol, trifluralin, ziram |
| Developmental or reproductive toxicants² | 2,4-Diethylamidine, benomyl, bromoxynil, carbafuran, cyazine, diazinon, diquat dibromide, s-ethyl dipropylthiocarbamate (EPTC), mancozeb, maneb, metat sodium, methyl bromide, methyl parathion, oxyfluorfen, propargite, s.s.s-trityl strut, triadimefon, vinclozolin |
| Organochlorides² | Dicofol, endosulfan, lindane |
| Organophosphates² | Acephate, azinphos-methyl, chlorpyrifos, diazinon, dimethoate, disulfoton, ethoprop, fonofos, malathion, methamidophos, methidathion, methyl parathion, mevinphos, naled, oxydemeton-methyl, parathion, phorate, phosmet, profenofos |
| Carbamates² | Nedidicarb, benomyl, carbyl, carbafuran, fenetur, methomyl, propoxide, propyzamide |
| Dithiocarbamates² | Mancozeb, maneb, metat sodium, thiram, zineb, ziram |

²Probable human carcinogens with sufficient evidence in laboratory animals and inadequate or no evidence in humans (10). ²Possible human carcinogens with limited evidence in laboratory animals (10). ²Positive in two or more laboratory assays (14,15). ²Positive in one or more developmental or reproductive studies in laboratory animals (16). ²Chemical groups were identified from Meister (17).
To prioritize individual pesticides for analysis, we developed a hazard weighting system based on two measures of carcinogenic potential and two measures of exposure potential. We assigned weights for each of these attributes to the highest-use pesticides from the toxicologic groups. The U.S. EPA cancer class was used to assign to each pesticide a weight from 1 through 10 based on the evidence that it is a carcinogen (10). Since there were no class A or class B1 carcinogens with geographically referenced use during our study period, the highest score assigned for cancer class was 7. Cancer slope factors, which estimate cancer potency from the dose–response relationship, have been calculated for all probable (class B2) and most possible (class C) carcinogens (10). As a second measure of carcinogenic potential, we assigned each pesticide a weight from 1 to 10 based on its cancer potency. If data were not available, a default weight of 1 was assigned to the pesticide for that attribute. Table 2 provides a key to the weights for each attribute.

We used volatilization flux rate and field half-life as measures of physical characteristics that could be associated with exposure potential. Volatilization flux estimates the tendency of a pesticide to move into the air after application and is correlated with the downwind concentration in air (18). We estimated the volatilization flux for each pesticide using the vapor pressure, water solubility, and soil absorption coefficient (19,20). Pesticides were assigned a weight from 1 through 10 on the calculated volatilization flux. We used the field dissipation half-life—a measure of the overall rate of disappearance of a pesticide from treated fields—as an indicator of persistence (20). Pesticides were assigned a weight from 1 through 5 based on persistence. The range used for persistence weight was half that used for volatilization flux weight because the dose received by children from ingestion of household dust is estimated to be about half the dose from inhalation for most pesticides (21,22). Moreover, we considered volatilization and secondary drift a necessary precursor for most potential exposures to children in nearby communities.

We calculated the cancer hazard factor for each pesticide by multiplying the weights for each attribute and then normalizing to make the highest possible score 10. The range of potential cancer hazard factors covers almost four orders of magnitude (0.002 to 10):

\[ \text{Cancer Hazard Factor} = \frac{\text{Cancer Potency} \times \text{Flux} \times \text{Persistence}}{500}. \]

We calculated hazard-adjusted pesticide use by multiplying the pounds applied by the corresponding cancer hazard factor. Individual pesticides were ranked by hazard adjusted use:

\[ \text{Hazard Adjusted Pesticide Use} = \frac{\text{Cancer Hazard Factor} \times \text{Pounds of Use}}{\text{Field-half-life}}. \]

Block-group exposure assessment. We used the 1991–1994 PUR data to calculate the annual average pesticide use in pounds for each square-mile section (23). We used the annual average because our focus was on cancer and chronic exposure. We used a GIS to determine the spatial relationship between sections and census-block groups. In 1990, California block groups had a median land area of 0.2 square miles and a huge range, from 0.001 to 3,610 square miles (24). Pesticide use was allocated from the section to each corresponding block group on the basis of percent area of the section in that block group. We calculated pesticide use density in pounds per square mile of census-block group by summing the average pounds applied in all relevant sections and then dividing by the block-group area. The median, 90th percentile, and maximum block-group pesticide use density were determined for each pesticide and pesticide group. We used 1990 census data to obtain the number of children under 15 years of age by block group. The number of children living in block groups with pesticide use density above the 90th percentile was calculated for each pesticide group and the highest cancer hazard ranking pesticides.

### Results

**PUR data.** For all pesticides reported in the PUR, the annual average agricultural pesticide use for 1991–1994 was greater than 169 million pounds. Correcting for application rates above the estimated maximum allowable rate reduced the average by 5% to 160 million pounds. Application rate errors were often an order of magnitude greater than the average rate, indicating data entry errors. Location errors further reduced statewide annual average pesticide use by another million pounds or less than 1%. The most frequent location error involved sections that were not within the reported county. Location errors occurred in more than 1,000 sections (0.5%) and affected a smaller number of pounds than high application rate errors. Given the size of the PUR database, we considered the observed error rate of approximately 6% of reported pounds relatively low.

**Pesticide use by groups.** The statewide annual average use for the pesticide groups is shown in Table 3. The probable and possible carcinogen groups each had about 10 million pounds per year of reported use, and the genotoxic and developmental/reproductive toxicant groups were both greater than 30 million pounds per year. Among the chemical classes, organochlorine insecticides had the least use with less than 1 million pounds per year, and the dithiocarbamate fungicides had the most use with greater than 10 million pounds per year.

To evaluate changes in pesticide use from 1991 to 1994, we graphed annual reported use for probable carcinogens, possible carcinogens, methyl bromide, and metam sodium (Figure 1). We chose methyl bromide and metam sodium because these were the highest use pesticides from the four toxicologic groups. The use of probable carcinogens (class B) increased from 8 to 16 million pounds from 1991 to 1994. Most of that increase was caused by metam sodium use, which grew from approximately 5 million to 11 million pounds. The largest increase occurred between 1991 and 1992.

### Table 2. Pesticide cancer hazard weights by attribute.

| Weight | Cancer class* | Cancer potencya (mg/kg/day) | Volatilization fluxb,c | Field-half-life* (days) |
|--------|---------------|-----------------------------|------------------------|------------------------|
| 10     | A             | > 1                         | > 10⁻¹                  | —                      |
| 8      | B1            | 0.1–1.1                     | > 10⁻²–10⁻¹            | —                      |
| 7      | B2            | —                           | > 10⁻³                  | —                      |
| 5      | C             | > 0.01–0.1                  | > 10⁻⁵–10⁻³            | > 100                  |
| 4      | G or D/Rd     | 0.001–0.01                  | 10⁻³–10⁻⁵              | 76–190                 |
| 3      | —             | 51–75                       | 26–50                  |
| 2      | NA            | < 0.001 or NA               | < 10⁻⁵ or NA           | < 25 or NA             |

NA, not available.

*aFrom U.S. EPA (10). bFlux rate = vapor pressure/(water solubility × soil absorption coefficient) from Glotfelty et al. (19). cVapor pressure, water solubility, soil absorption, and field half-life from U.S. Department of Agriculture (20). dGenotoxic or developmental/reproductive toxicant (18).

### Table 3. Average annual pesticide use in California from 1991 to 1994 for pesticide groups.a

| Pesticide group | Average poundsa |
|-----------------|-----------------|
| Class B carcinogens | 12,643,173 |
| Class C carcinogens | 9,372,325 |
| Genotoxic compounds | 38,445,168 |
| Developmental/reproductive toxicants | 31,472,459 |
| Organochlorines | 903,550 |
| Organophosphates | 6,687,806 |
| Carbamates | 2,326,545 |
| Dithiocarbamates | 10,884,652 |

*aIndividual pesticides can be in more than one group.

PUR data corrected for erroneously high application rates and includes only valid geographic locations.
which may reflect increased awareness of the legal mandate of reporting (12). This time period also coincides with severe restrictions on the use of 1,3-dichloropropene (Telone), a fumigant that was largely replaced by metam sodium. The use of possible carcinogens (class C) and methyl bromide remained relatively constant.

**Pesticide cancer hazard prioritization.**

The calculated cancer hazard factors for individual pesticides (Table 4) ranged over more than two orders of magnitude, although most pesticides had hazard factors between 0.1 and 1.0. For pesticides classified as probable or possible carcinogens, the cancer hazard weights are greater than the exposure potential weights because of the lesser weighting for persistence. The cancer hazard factors for pesticides from the other toxicologic groups were more influenced by their exposure potential.

The relative ranking of pesticide use changed significantly when pounds were adjusted by the cancer hazard factors. The top pesticides in the state ranked by hazard-adjusted use (Table 5) were propargite, methyl bromide, and trifluralin. The top pesticides from the toxicologic groups ranked by pounds alone were methyl bromide, metam sodium, and chlorpyrifos. Propargite had a larger cancer hazard factor than some high-use pesticides, such as chlorpyrifos, producing a much higher ranking by hazard-adjusted use.

**Block-group exposure assessment.**

We calculated the statewide distribution of pesticide use density among block groups with more than 1 pound per square mile of use for a given pesticide group or individual pesticide (Table 6). Very low pesticide use densities may have been the result of location errors within counties that could not be eliminated. Therefore, we considered block groups with use densities less than 1 pound per square mile to have little potential exposure. There were 3,000–9,000 census-block groups in the state with more than 1 pound per square mile of pesticide use for each pesticide group. The median densities were generally greater than 10 pounds per square mile. The distributions were not normal with order-of-magnitude increases between the median, 90th percentile, and maximum use densities. The 90th percentile of use density was around 500 pounds per square mile for the two carcinogen groups and greater than 1,500 pounds per square mile for the genotoxic and developmental or reproductive toxicant groups. Among the chemical classes, organochlorines had the lowest use density and dithiocarbamates had the highest, with a median of 30 pounds per square mile.

For individual pesticides, the number of block groups with more than 1 pound per

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**Figure 1.** Annual agricultural pesticide use in California from 1991 to 1994. Metam sodium is included among Class B carcinogens. PUR data corrected for erroneously high application rates and includes only valid geographic locations.

**Table 4.** Cancer hazard weights and factors for pesticides in toxicologic groups with annual use > 150,000 pounds per year.

| Pesticide                     | Cancer class weight | Cancer potency weight | Volatilization flux weight | Field half-life weight | Cancer hazard factor |
|-------------------------------|--------------------|-----------------------|---------------------------|------------------------|---------------------|
| Probable carcinogens          |                    |                       |                           |                        |                     |
| Captan                        | 7                  | 3                     | 5                         | 1                      | 0.210               |
| Chlordaneol                   | 7                  | 3                     | 8                         | 2                      | 0.672               |
| Ipodione                      | 7                  | 5                     | 5                         | 1                      | 0.350               |
| Mancozeb                      | 7                  | 5                     | 1                         | 2                      | 0.140               |
| Maneb                         | 7                  | 5                     | 1                         | 2                      | 0.140               |
| Metam sodium                  | 7                  | 8                     | 1                         | 1                      | 0.112               |
| Propargite                    | 7                  | 5                     | 5                         | 4                      | 1.400               |
| Possible carcinogens          |                    |                       |                           |                        |                     |
| Acephate                      | 5                  | 5                     | 3                         | 1                      | 0.150               |
| Carbarly                      | 5                  | 5                     | 5                         | 1                      | 0.250               |
| Chlordih-dimethyl             | 5                  | 3                     | 8                         | 3                      | 0.720               |
| Cyanazine                     | 5                  | 10                    | 3                         | 2                      | 0.600               |
| Dicofol                       | 5                  | 8                     | 5                         | 3                      | 1.200               |
| Dimethoate                    | 5                  | 1                     | 5                         | 1                      | 0.050               |
| Fosetyl-al                    | 5                  | 1                     | 1                         | 1                      | 0.010               |
| Methidathion                  | 5                  | 1                     | 8                         | 1                      | 0.080               |
| Metolachlor                   | 5                  | 1                     | 8                         | 5                      | 0.400               |
| Moline                        | 5                  | 8                     | 10                        | 1                      | 0.800               |
| Nonflurazone                  | 5                  | 1                     | 5                         | 5                      | 0.250               |
| Drycain                       | 5                  | 8                     | 5                         | 1                      | 0.400               |
| Oxylflurafen                  | 5                  | 8                     | 5                         | 2                      | 0.800               |
| Pendimethacil                 | 5                  | 1                     | 8                         | 5                      | 0.400               |
| Permethrin                    | 5                  | 5                     | 5                         | 2                      | 0.500               |
| Propham                       | 5                  | 1                     | 5                         | 1                      | 0.250               |
| Simazine                      | 5                  | 8                     | 5                         | 4                      | 1.600               |
| Trifluralin                   | 5                  | 3                     | 10                        | 4                      | 1.200               |

*Genotoxic or developmental/ reproductive toxicants*

| Pesticide          | Cancer class weight | Cancer potency weight | Volatilization flux weight | Field half-life weight | Cancer hazard factor |
|--------------------|---------------------|-----------------------|---------------------------|------------------------|---------------------|
| 2,4-Diethylanlaine | 3                   | 1                     | 5                         | 2                      | 0.060               |
| Aldicarb           | 3                   | 1                     | 5                         | 2                      | 0.060               |
| Chlorofuran        | 3                   | 1                     | 5                         | 2                      | 0.060               |
| Chloropicrin       | 3                   | 1                     | 10                        | 1                      | 0.060               |
| Chlorpyrinos       | 3                   | 1                     | 8                         | 1                      | 0.060               |
| Diazinon           | 3                   | 1                     | 5                         | 1                      | 0.030               |
| Ethyl dipropylthiocarbamate | 3 | 1     | 10                        | 1                      | 0.060               |
| Malathion          | 3                   | 1                     | 5                         | 1                      | 0.030               |
| Methyl bromide     | 3                   | 1                     | 10                        | 2                      | 0.120               |
| Mivemphos          | 3                   | 1                     | 5                         | 1                      | 0.030               |
| Paraquat dichloride| 3                   | 1                     | 10                        | 1                      | 0.030               |
| S,S,S-triethyl      | 3                   | 1                     | 5                         | 2                      | 0.060               |
| Ziram              | 3                   | 1                     | 5                         | 2                      | 0.060               |

*Cancer hazard factor = (evidence weight × potency weight × flux weight × persistence weight)/500. *Not available.*
square mile of use varied tremendously from 194 for molinate to $>3,400$ for methyl bromide. The 90th percentile of use density was greater than 100 pounds per square mile for most individual pesticides. The soil fumigants methyl bromide and metam sodium had much higher use densities than the other individual pesticides with 90th percentile values greater than 1,500 pounds per square mile.

To illustrate the methods used to calculate block-group pesticide use density, an example is provided from Fresno, California. Figure 2A shows probable carcinogenic pesticide use in pounds by section and Figure 2B shows the resulting use density in pounds per square mile for census-block groups in this area. The block-group pesticide use density essentially follows the section-level pesticide use. Figure 2B also illustrates that larger, rural block groups tend to have the highest pesticide use density and smaller, urban block groups the lowest. In high-use rural areas, census-block group mapping is less geographically specific than mapping by section because of the large area of these block groups.

We mapped the geographic distribution of pesticide use density by block group using the percentiles of the statewide distribution for all probable carcinogens (Figure 3) and for propargite, which was the highest-ranking individual compound (Figure 4). For probable carcinogens, the highest use areas were in the San Joaquin, Sacramento, Salinas, and Imperial Valleys. This corresponds well with the heaviest agricultural counties in the state based on farm revenues (25). Propargite use was not as geographically widespread, and the high-use density area was primarily the San Joaquin Valley.

More than 6.6 million children under 15 years of age lived in California in 1990. The number of children living in block groups above the 90th percentile of use density varied considerably among the pesticide groups and individual pesticides (Table 6). Developmental or reproductive toxicants had the most children with nearly 267,000, and molinate had the least number of children with just over 3,300. Organophosphates and organochlorides had about 200,000 and 60,000 children living in these high-use block groups, respectively. The variation in the number of children living in these block groups demonstrates that different populations were potentially exposed for each group and individual pesticide.

**Table 5.** Highest-ranking pesticides based on hazard-adjusted use, 1991–1994.

| Pesticide     | Cancer hazard factor | Corrected pounds<sup>a</sup> | Hazard-adjusted use<sup>b</sup> |
|---------------|----------------------|------------------------------|---------------------------------|
| Propargite    | 1.400                | 1,600,982                    | 2,241,375                       |
| Methyl bromide| 0.120                | 16,901,451                   | 2,028,174                       |
| Trifluralin   | 1.200                | 1,230,218                    | 1,476,262                       |
| Simazine      | 1.600                | 869,962                      | 1,391,939                       |
| Molinate      | 0.800                | 1,380,424                    | 1,104,339                       |
| Metam sodium  | 0.112                | 8,300,569                    | 929,664                         |
| Dicofol       | 1.200                | 704,077                      | 664,892                         |
| Chlorothalonil| 0.672                | 786,572                      | 528,576                         |
| Chlorothal-dimethyl| 0.720            | 642,891                      | 462,882                         |
| Oxyfluorfen   | 0.600                | 334,325                      | 267,460                         |
| Oryzalin      | 0.400                | 667,445                      | 266,978                         |
| Cyanazine     | 0.600                | 411,331                      | 246,769                         |
| Chlorpyrifos  | 0.096                | 2,429,610                    | 233,243                         |
| Carbaryl      | 0.250                | 820,487                      | 205,122                         |
| Iprodione     | 0.350                | 408,562                      | 142,997                         |
| Chloropicrin  | 0.060                | 2,364,831                    | 141,890                         |
| Pendimethalin| 0.400                | 204,845                      | 113,938                         |
| Permethrin    | 0.500                | 201,795                      | 100,898                         |
| Ziram         | 0.080                | 1,590,812                    | 95,449                          |
| Captan        | 0.210                | 417,612                      | 87,699                          |

<sup>a</sup>Hazard adjusted use = corrected pounds × cancer hazard factor.

**Table 6.** Distribution of annual average agricultural pesticide-use density in California census-block groups for toxicologic groups, chemical groups, and high-hazard individual pesticides.<sup>c</sup>

| Toxicologic groups       | Block groups<sup>a</sup> | Median (lbs/m<sup>2</sup>) | 90th percentile (lbs/m<sup>2</sup>) | Max (lbs/m<sup>2</sup>) | Children (< 15 years) in 90th percentile<sup>c</sup> |
|--------------------------|--------------------------|---------------------------|------------------------------------|-------------------------|-----------------------------------------------|
| Class B                  | 4,932                    | 31                        | 569                                | 14,935                  | 169,884                                       |
| Class C                  | 6,218                    | 23                        | 445                                | 5,043                   | 198,375                                       |
| Genotoxic                | 7,505                    | 48                        | 1,844                              | 70,670                  | 261,333                                       |
| Developmental/reproductive | 6,647                  | 45                        | 1,789                              | 48,784                  | 266,960                                       |
| Chemical groups          |                          |                           |                                    |                         |                                               |
| Organochlorides          | 3,881                    | 9                         | 569                                | 60,909                  |                                               |
| Organophosphates         | 9,288                    | 18                        | 249                                | 7,129                   | 204,144                                       |
| Carbamates               | 6,755                    | 14                        | 141                                | 1,706                   | 138,316                                       |
| Dithiocarbamates         | 3,216                    | 14                        | 764                                | 14,931                  | 109,474                                       |
| Individual pesticides    |                          |                           |                                    |                         |                                               |
| Propargite               | 2,144                    | 21                        | 172                                | 926                     | 61,892                                        |
| Methyl bromide           | 3,421                    | 163                       | 2,688                              | 46,105                  | 127,562                                       |
| Trifluralin              | 1,287                    | 14                        | 118                                | 784                     | 35,983                                        |
| Simazine                 | 2,109                    | 15                        | 112                                | 582                     | 64,462                                        |
| Molinate                 | 194                      | 49                        | 696                                | 1,433                   | 3,334                                         |
| Metam sodium             | 1,072                    | 86                        | 1,503                              | 14,480                  | 42,145                                        |
| Dicofol                  | 1,342                    | 7                         | 72                                 | 352                     | 44,902                                        |
| Chlorothalonil           | 2,369                    | 13                        | 109                                | 2,537                   | 84,740                                        |

<sup>c</sup>Calculated from census-block groups with use density $>1$ lb/m<sup>2</sup> for that pesticide. <sup>b</sup>Number of block groups with $>1$ lb/m<sup>2</sup> use density for that pesticide; total block groups used in this analysis were 21,443. <sup>c</sup>Number of children under 15 years of age living in census-block groups above the 90th percentile of pesticide-use density.
counterparts. Population growth in California has led to the development of suburban areas adjacent to fields or on former farmland, increasing the potentially exposed population.

We consider pesticide use density an indicator for a wide range of potential exposure pathways, including inhalation of ambient air, soil drift and persistence in household dust, potential groundwater contamination, parental occupational “take home” exposures, playing in fields, and eating produce directly from treated fields.

Hazard-weighted pesticide use created different priorities for assessing individual compounds (Table 5). Our focus was on ranking carcinogens for a childhood cancer study, but these hazard-weighting methods could be modified for other health outcomes of interest (37, 38). A hazard scoring system used by the Department of Pesticide Regulation to evaluate pesticides as toxic air contaminants also ranked propargite, simazine, chlorothalonil, molinate, metam sodium, cyanazine, and chlorpyrifos among the top 20 compounds (39). Methyl bromide, trifluralin, carbaryl, and captan are already classified as toxic air contaminants in California pursuant to section 14021(b) of the Food and Agricultural Code (39).

Nonoccupational exposures to molinate are suggested to exceed safety margins (40). Methyl bromide, chlorothalonil, and molinate have been detected in ambient air of agricultural communities in California (27).
There are some notable limitations to weighting pesticide use by cancer hazard. Pesticides that have not been toxicologically tested for carcinogenicity, genotoxicity, or developmental/reproductive toxicity were not included in our prioritization. Eleven pesticides with more than 1 million pounds per year of use in California had insufficient toxicologic and environmental data for hazard weighting (sulfur, petroleum oil, sodium chloride, copper hydroxide, mineral oil, copper sulfate, chloropicrin, petroleum distillates, sulfuric fluoride, calcium hydroxide, and diuron). Furthermore, the weighting of each hazard attribute and exposure relative to carcinogenicity may not reflect true environmental and biologic activity. Animal cancer potency may not accurately reflect the potency for humans, although the evidence is fairly convincing that human carcinogens are carcinogenic in rodents (41). Some pesticides degrade into compounds that have more or less carcinogenic or exposure potential than the original parent compound. For example, the actual fumigant action of metam sodium (a probable carcinogen) comes from a reaction product called methyl isothiocyanate, which is not thought to be a carcinogen. Information on the environmental breakdown products of pesticide active ingredients was not included in our prioritization system because data were not available for most pesticides.

The PUR system has some limitations that are potentially problematic for epidemiologic studies. Information on residential pesticide use in the home and garden is not collected. Agricultural pesticide use is reported to a square-mile section, but air monitoring data from application sites suggest that pesticide concentrations may decrease significantly within a mile (18,42–44). Nonagricultural pesticide applications, including structural fumigations and landscaping uses on golf courses and along highways, are reported only at the county level in the PUR database. Improved spatial resolution for both agricultural and structural/landscaping applications would represent a significant refinement to the PUR system for use in health studies. The PUR system is legally mandated, but pesticide use is self-reported, and underreporting has not been evaluated. Information on the type and amount of inert ingredients applied is not provided. Many of the solvents used in pesticide formulations also have toxicologic effects of concern (45,46). Despite these limitations, the PUR system is still probably the most comprehensive agricultural pesticide use database in the world (12).

We calculated the annual average pesticide use density to examine chronic exposure. However, pesticide applications are frequently seasonal, and many are applied only once per year or in response to specific pest infestations. If the PUR data are to be used for studies of other health outcomes, the relevant time period should be considered. Because of the geographic resolution of the PUR data, we assumed that pesticide use was distributed evenly within a square-mile section. Pesticide use density represents pesticide use averaged over the entire land area of the block group, but all applications could have occurred in a single section.

The PUR data represent an extremely valuable resource for conducting health studies. Residents are unlikely to have knowledge about pesticide use on nearby fields, unlike home and garden use. The measures presented here are based on independent reporting and do not rely on recall by study participants. The PUR data also allowed for evaluation of specific pesticide active ingredients and the combination of pesticides with similar chemical or toxicologic properties. For other health studies, pesticide groups should be tailored to the health outcomes or exposure pathways of interest. A GIS was essential in conducting this analysis because it allowed for the spatial overlay of agricultural pesticide use and census-block groups.

The heavy use of potentially toxic agricultural pesticides in some areas of California warrants further exposure and epidemiologic investigation. Environmental and biologic monitoring is needed to determine the relationship between agricultural pesticide use and individual exposure. Additional toxicologic data are also desirable for high-use pesticides. The range of values reported here for census-block group pesticide use density are suitable for a statewide epidemiologic study of childhood cancer. The number of children living in both high and low pesticide-use density areas is sufficient to allow for the spatial overlay of agricultural pesticide use and census-block groups.

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