

SUPPORTING INFORMATION (SI):

Pesticides in Dust from Homes in an Agricultural Area

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TABLE OF CONTENTS

<u>Pesticide Use Reporting (PUR) data for the Salinas Valley</u>	3
<u>CHAMACOS Study Population</u>	4
<u>Home-Visit</u>	4
<u>Geographic Positioning System Readings</u>	4
<u>Pesticide Use Near Homes</u>	4
<u>Weather Information</u>	5
<u>Chemical Analysis:</u>	5
GC/MS Method:	6
LC/MS /MS	7
GC/MS Method Performance	7
Analyte Detection in GC/MS Method	8
<u>Statistical Analysis:</u>	9
Selection of Samples from Separate Residences	9
Selection of Independent Variables	9
Regression Analysis	10
<u>Mapping of Pesticide Use and Dust Concentrations:</u>	10
 Table S.1: Average Annual kg of Pesticides Used Agriculturally in the Salinas Valley in 1999-2002	11
Table S.2: Pesticide Use in Salinas Valley and Environmental Fate Physicochemical Parameters of Dust Analytes	12
Table S.3: QC results for Spiked Dust and Duplicate Dust Analyses	13
Table S.4: Average and distribution of SRS recoveries in 55 QC samples and 504 field samples by GC/MS	14
Table S.5: Detection Frequency and Concentration Range by different ranges of dust mass extracted	15
Table S.6: Bivariate Regression Results: Proportional Change in Dust Concentrations for Unit Change in Predictor Variables	16
 Figure S.1a and 1b: Chlorpyrifos use and dust concentrations in the Salinas Valley	18
Figure S.2a and 2b: Diazinon use and dust concentrations in the Salinas Valley	19
Figure S.3a and 3b: DCPA use and dust concentrations in the Salinas Valley	20
Figure S.4a and 4b: Iprodione use and dust concentrations in the Salinas Valley	21
Figure S.5a and 5b: Permethrin use and dust concentrations in the Salinas Valley	22

Pesticide Use Reporting (PUR) data for the Salinas Valley: In California, all pesticide applications to agricultural fields are required to be reported by the pesticide user or property “operator.” Operators are required to use a standardized form (the PUR report) (1) to report the EPA registration number of the pesticide product applied, amount used, acres treated, date and time applied, method of application (air, ground, or other), location of application, and the name of the pesticide applicator. Location is recorded according to the Public Land Survey System (PLSS) which divides the State into sections of approximately 1 mile² (2.6 km²).

Pest control businesses, public agencies, and other persons are also required to report several other non-agricultural types of applications on a different standardized form. These “non-agricultural” applications include: structural, landscape maintenance, right-of-way, public health, vertebrate, commodity fumigation, and regulatory pest control (2). For structural and landscape maintenance, licensed pest control operators are required to report all amounts applied. Other California residents, however, are not required to report home and garden use or institutional use. Although the county where the pesticides are applied for these other non-agricultural applications is reported, geographic location is *not* further specified.

Both agricultural and non-agricultural pesticide uses are reported to the County Agricultural Commissioner’s office who then reports it to the California Department of Pesticide Registration (CDPR) of the California Environmental Protection Agency (EPA). CDPR compiles the pesticide use data from all of the 58 counties in California, conducting up to 50 different validity checks. CDPR’s data processing includes matching the U.S. EPA registration number for the pesticide product applied with the active ingredients, or pesticides, in each product. CDPR flags a use record as an outlier if it meets any one of three criteria: 1) applications over 200 pounds of active ingredient per acre; 2) applications over 50 times the median application rate for the crop and product treated; and 3) applications defined by a group of 12 scientists as obvious outliers. Data is verified and checked with the County Agricultural Commissioner’s office. After the data verification process, the CDPR produces a summary of the pesticide use in each year. An online-query system is available where agricultural and non-agricultural pesticide use by year, county, and commodity can be assessed. In addition, zip files of the PUR dataset for all applications (agricultural and non-agricultural) for each year, with all items on the agricultural PUR form, including the PLSS section of application, can be downloaded or requested (3,4).

We obtained the PUR dataset from CDPR for the years within which samples were collected (1999, 2000, and 2001) for all of California. To summarize agricultural pesticide use in the Salinas Valley (SV), the region was first defined as an area bounded by the ocean and an isopleth at 60 m in elevation. As defined, the SV included the agricultural areas of Monterey County and relatively small portions of San Benito and Santa Cruz Counties. Specifically, the SV was 83%, 13%, and 3% Monterey, Santa Cruz, and San Benito Counties, respectively. We identified the PLSS sections within the SV and abstracted the agricultural pesticide use within the SV from the California PUR dataset. For agricultural applications in 1999-2001 in the SV, 0.6-2% of pounds of pesticides applied in the SV had been flagged by CDPR as outliers. We replaced these outliers with the county, crop, and product-specific median pounds of active ingredient applied per application. The corrected SV PUR dataset was used to calculate average annual agricultural pesticide use in the SV.

Amounts of pesticides used agriculturally in the SV are given in Table S.1 and Table S.2. In addition, other non-agricultural pesticide uses of target analytes were estimated for the SV from the amounts reported for each of the three counties in the SV and are given in Table S.2. For diazinon, chlorpyrifos, and permethrin, the non-agricultural uses in Monterey County are 2 to 3% of the agricultural use in the SV, but for the other analytes, the non-agricultural uses were less than 1% of the agricultural use.

CHAMACOS Study Population: Women were eligible to participate if they were ≤ 20 weeks gestation at the time of enrollment, were eighteen years or older, and qualified to receive poverty-based government health insurance. Informed consent was obtained from all participants in accordance with procedures approved by the University of California Berkeley Committee for the Protection of Human Subjects (5,6). Participants' and their babies had study visits at the clinic at enrollment (≈ 13 weeks gestation) and when the baby was six and twelve months old. At the clinic visits, demographic, health, and household information was collected through personal interviews with the mother. Interviews were conducted in English or Spanish by bilingual and bicultural staff. Household information included pesticide use in the home in the past six months and occupation, which included whether they worked with pesticides and whether they worked in the fields in the past two months, or since the last visit.

Home-Visit: Detailed home inspections were conducted at the enrollment visit, and when the child was six and twelve months old. Recorded information included how many people lived in the home and, of those, how many worked as farmworkers or in agriculture, and the approximate distance from the home (i.e., < 200 feet (60 m) to a quarter mile (400 m)) to the closest agricultural field or orchard. Overall quality of the housekeeping or "cleanliness" was rated on a three point scale according to the amount of grease around stoves, the presence of dirty dishes and overflowing trash, and the presence of dust, dirt, and food particles on floors and behind cabinets, appliances, or furniture (7).

The labels of any home pesticide products were read and the active ingredients and the US EPA registration numbers were recorded. After the home-visit, the US EPA registration was checked using an online database to confirm the recorded active ingredients.

Geographic Positioning System Readings: Geographical positioning system (GPS) readings were recorded at the home visit. When the participant had not moved between two visits, the two GPS readings were compared, and if they were more than 15 m off, recordings were verified with aerial overlay maps. If the discrepancy was not resolved, a visit to the house was made and an additional GPS reading recorded.

Pesticide Use Near Homes: To sum the PUR near the participant's home for use as independent variables in regression analysis, we used the longitude and latitude obtained by GPS reading during each home visit to identify the PLSS within which the home was located. Because the participant could live anywhere in a PLSS section, including near the border, we

summed the agricultural PUR use “near” the home including the use in the PLSS section in which the participant lived and the eight sections surrounding that PLSS section (for a total area of ≈ 9 miles² or 23 km²). We used the corrected SV PUR dataset as described above and summed the use in three mutually exclusive, lagged periods: days (days 1-3 prior to sample collection), month (days 4-33 prior to collection) and season (days 34-133 prior to sample collection). Granular applications (approximately 25% of the kg applied) were not included in the OP PUR sums, as granular products have lower volatility.

Weather Information: GPS readings at the home visits were matched to one of two weather stations in the SV(8). Daily temperature and rainfall information was then matched and averaged for the periods corresponding to the lagged periods for the PUR sums.

Chemical Analysis: All dust samples were stored on ice packs in the field and during transport to the field laboratory, where they were stored at -80°C. The selected 504 samples were shipped to the analytical laboratory (Battelle Memorial Institute, Columbus, Ohio), where they were stored at -20°C until analysis.

Each dust sample was sieved to <150 μ m, and the mass of the fine dust (0.5-150 μ m) was recorded. Where feasible, two 0.5 g aliquots were weighed out for the two separate chemical analyses. If the total dust amount was 0.05 – 0.99 g, then the dust was split into two equivalent amounts for analysis. When the dust amount was <0.05 g, the total amount was dedicated to the analysis method that encompassed the larger number of pesticides. Each method involved solvent extraction by ultrasonication, chromatographic cleanup, and analysis with a mass spectrometry technique. Gas chromatography/mass spectrometry (GC/MS), in the multiple ion detection mode (MID), was used for detection of 17 pesticides. High performance liquid chromatography coupled to electrospray tandem mass spectrometry (LC/MS/MS) was used for detection of five pesticides that were either very polar, and thus not amenable to GC separation, or thermally labile, and thus readily degraded in the hot GC injector.

Any multi-residue extraction and cleanup method is taxed when trying to encompass analysis of both very low polarity organochlorines and very high polarity, small organophosphorous insecticides (OPs) (e.g., acephate and dimethoate), and other classes of intermediate polarity. To track extraction/cleanup method performance on a sample-by-sample basis, we added compound class-specific surrogate recovery standards (SRSs). The SRSs included: ¹³C₆-*trans*-permethrin and ¹³C₆-*cis*-permethrin (to track method performance for relatively non-polar analytes), fenchlorphos (to track performance for moderately polar OPs), and ¹³C₁-diethyacetamidomalonate (DEAA) (to track high polarity OPs). In addition, ¹³C₁₂-*p,p'*-DDE was added specifically to correct for planned losses of *p,p'*-DDE. The cleanup step to remove aliphatic hydrocarbons and lipids from the extract resulted in about 80% loss of the *p,p'*-DDE from the extract; this SRS facilitated an isotope dilution type of quantification for this one analyte. Though structurally similar, *p,p'*-DDT and chlorthal-dimethyl (DCPA) did not elute as early in the cleanup step as *p,p'*-DDE.

Samples were extracted and analyzed in 33 batches of 16 field samples, one solvent method blank, and two additional QC samples. For the GC/MS method, the two additional QC samples per batch rotated between: i) a solvent spike (100 ng/analyte) and a samples duplicate, ii) a reference dust and the reference dust spiked at a low level (50 ng per analyte), and iii) the reference dust and the reference dust spiked at a high level (500 ng per analyte). For the LC/MS/MS method, the two additional QC samples per batch always included a sample duplicate, and the last QC sample rotated between: i) the reference dust, ii) the reference dust spiked at a low level (25 ng per analyte), and iii) the reference dust spiked at a high level (250 ng). The reference dust was obtained from a vacuum cleaner bag from a central Ohio home; this dust was stored as a sieved bulk dust (>500 g) at room temperature. Since multiple aliquots of the dust were sieved over time, no attempt was made to standardize the dust; each aliquot used as a QC sample was treated as an individual sample.

GC/MS Method: All dust and QC samples were spiked with 250 ng of the 5 SRSs listed above. Each dust was extracted in 1:1 hexane:acetone for 10 minutes by ultrasonication; the solvent of the resulting extract was exchanged into hexane prior to cleanup on silica. The solid phase extraction (SPE) cleanup on silica (1 g, BakerBond) included sequential elution with hexane, 15% diethyl ether in hexane, dichloromethane (DCM), and 20% acetone in ethyl acetate. The hexane fraction was discarded. The internal standard (IS) for quantification, dibromobiphenyl, was added to the final 1 mL extract prior to GC/MS analysis.

Seven calibration curve solutions (plus a 0 level solution), covering a concentration range of 2-500 ng/mL, were analyzed together with each sample set using an Agilent/HP 6890 GC interfaced to an Agilent/HP 5973 MSD. Extracts were analyzed using a 30 m DB1701 column, with a temperature program of 70 -130 °C @25 °C/min, 130-220 °C @2 °C/min, and 220-280 °C @ 10 °C/min. At least two ions (quantification and qualifier) were monitored for each analyte. Identification was based on three criteria: correct retention time, co-maximized peak shapes for quantification and qualifier ions, correct peak area ratio of quantification ion to qualifier ion.

Linear regression analysis was used to establish the calibration curve for each analyte in each sample batch. For dust sample extracts where the concentration of one or more analytes exceeded the calibration curve maximum concentration by 15%, the sample extract was diluted, re-spiked with IS, and reanalyzed.

For data analysis, sample concentrations were adjusted by the recovery in that sample of the matched SRS as follows: fenchlorphos (azinphos methyl, chlorpyrifos, diazinon, fonofos, malathion, methidathion, phosmet, vinclozoline); ¹³C₆-*trans*-permethrin (chlorthal dimethyl, *p,p'*-DDT, iprodione, *trans*-permethrin); ¹³C₆-*cis*-permethrin (*cis*-permethrin), and ¹³C₁-DEAA (acephate, chlorpyrifos oxon, dimethoate). In two samples, labeled *cis*-permethrin was not detected, and the recovery of labeled *trans*-permethrin was used to correct the recovery of native *cis*-permethrin.

LC/MS/MS: Pesticides that were not amenable to GC/MS, either because of polarity or sensitivity to heated GC injection, were analyzed using LC/MS/MS. The extraction method was driven by oxydemeton methyl which is temperature sensitive and degrades even with relatively low temperatures used in solvent concentration steps. A 10 mL aliquot of 9:1 hexane:acetone was added to the dust aliquot and the pesticides were extracted by ultra-sonication for 10 min. A 1 mL aliquot was removed, flushed with a stream of dry nitrogen to concentrate the extract to 0.8 mL. A 1.6 mL aliquot of acetonitrile saturated with hexane, which contained the IS for quantification ($^{13}\text{C}_1$ -DEAA), was added, and the sample was partitioned by shaking for 30 sec. A 1 mL aliquot of the acetonitrile layer was drawn off and filtered through a 0.45 μm syringe filter (Acrodisc 13CR, PTFE).

A six point calibration curve spanning the range of 0.1-100 ng/mL, plus a 0 level solution, was analyzed with each sample batch. Samples were analyzed using a Waters Alliance 2690 HPLC pump interfaced to a Micromass Quattro tandem mass spectrometer (MS/MS) that was operated in the positive ion electrospray mode. Two mass transitions (mass_1 to mass_2) were monitored for each analyte. Extracts were separated on an Agilent Zorbax RX-C18 HPLC column (2.1 x 150 mm), with a mobile phase constituents of 0.01 M formic acid in acetonitrile (A) and 0.01 M aqueous formic acid, programmed from 15:85 A:B to 85:15 A:B in a linear ramp over 19 minutes. All quantification methods were similar to those described above for the GC/MS analyses.

GC/MS Method Performance: The SRSs were added to each sample prior to extraction for two reasons. First, SRSs were added to track and assess method performance on a sample-by-sample basis by general compound class polarity range, since any multi-residue method is likely to suffer some deficiencies in trying to cover the broad range of compound classes represented here. Method performance is especially important for the cleanup method, as the wider the compound class polarity range of interest, the more co-extracted matrix material will be retained through cleanup, and this excess matrix material can affect detection capability of compounds where relatively low mass ions (i.e., $m/z < 150$) are monitored for detection (e.g., for malathion, acephate, azinphos methyl).

Second, in a manner similar to an isotope dilution method (where a chemically labeled version of each analyte is added prior to extraction, and quantification of the native analyte is automatically corrected by recovery of the isotope), these SRSs were added to expand the concept of isotope dilution analysis and allow for correction of analytical losses. An isotope dilution method requires a labeled analog for each analyte and the availability and cost of individual labeled analogs makes isotope dilution an unacceptable approach for an analysis with more than a few analytes. For this reason, we chose a limited number of SRSs, representing the polarity ranges of the analytes. This tracking of method performance by polarity was expected to be important because cleanup on silica can be variable. Specifically, both the activity of silica, and adverse impacts on the silica activity can occur if any acetone remains after the solvent exchange step (remaining acetone will deactivate the silica and cause analytes to elute early, in the discarded hexane fraction). This method represents the first time that the concept of isotope dilution analysis has been expanded in this way and applied to a multi-residue method where more than a few selected analytes are being monitored.

For this approach to be successful, the recovery of each SRS had to be similar to the recovery of the matched analytes. The SRSs were generally well-matched in method performance to the specific target analytes in that average polarity class recoveries (Table S.3) were within 15% of the SRS recoveries (Table S.4). At the higher spike level, where recovery data are available for all analytes, the average recovery for each SRS and the average uncorrected recoveries of matched analytes were: $^{13}\text{C}_6$ -*cis*-permethrin ($80 \pm 20\%$) and matched analytes ($95 \pm 19\%$); $^{13}\text{C}_6$ -*trans*-permethrin ($83 \pm 20\%$) and matched analytes ($86 \pm 6\%$); fenchlorphos ($84 \pm 15\%$) and matched analytes ($94 \pm 9\%$); $^{13}\text{C}_1$ -DEAA ($86 \pm 13\%$) and matched analytes ($81 \pm 20\%$). These results, though, have indicated that more SRSs need to be added to more precisely match the compound classes being investigated, and this has been done successfully in further studies (data not shown).

The solvent method blanks, with minor low-level exception, had no detectable pesticides; the two pesticides detected in solvent method blanks with greatest frequency were *cis*- and *trans*-permethrin (11 and 4 times, respectively, out of 38 blanks). These detections were due to low level impurity of native material in the labeled SRS. Data for dust samples were not corrected by solvent method blank levels because averages over 38 blank samples were less than detection limits.

The SRS-corrected recoveries of pesticides in the spiked reference dust samples are listed in Table S.3, together with the average relative percent difference (RPD) for detected analytes in sample duplicates; this average RPD only includes sample detects and does not include the 0% RPD for non-detects in samples. The recoveries for spikes into solvent were generally higher than dust spike recoveries (data not shown).

The average recoveries of the SRSs in the QC samples (blanks, spikes and duplicates) and in the 504 field samples are listed in Table S.4. Average recoveries in the two types of samples are quite similar ($>80\%$), indicating that the method performance indicated by the QC samples would be expected for the analyses of the field samples. Although average SRS recoveries were quite acceptable ($>80\%$), approximately one third of samples had SRS recoveries with less than the desired 75%. The distribution of SRS recoveries in field samples into higher than desired, acceptable, and lower than desired recoveries are also shown in Table S.4. These lower than desired SRS recoveries occurred in whole sample sets, indicating that a systematic error probably occurred in the solvent exchange step prior to SPE cleanup.

Analyte Detection in GC/MS Method: Quantification Limits (QL) were determined as the concentration yielding 10:1 signal:noise in a calibration curve solution, and did not include expected variation introduced by the dust matrix. The Limit of Detection (LOD) was the concentration yielding 3:1 signal:noise. Because we did not scale the final solvent volume to the dust mass extracted, for data evaluation purposes, we grouped samples into three levels of dust mass extracted (0.15-0.50 g; 0.05-0.14 g, and 0.01-0.04 g) to assess detection. We did this for two reasons: first, the lower dust mass in each range facilitated analyte detection, as there was not as much competing matrix material in the baseline signal; second, the ~3-fold difference in mass within a level is matched by a 3-fold difference between QL and LOD (e.g., an LOQ of 10

ng/mL for a 0.5 g sample equals 20 ng/g, and an LOD of 3 ng/mL for 0.15 g also equals 20 ng/g). The QL for each analyte in each of the three mass ranges is listed in Table S.5, together with the frequency of detection in each mass range, and the range of detected concentrations in each level. As expected, detection frequency decreased with successively lower amounts of dust extracted, and in cases such as chlorpyrifos and diazinon, the ~10X reduction in mass was matched by an ~10X reduction in detection frequency.

Statistical Analysis:

Selection of Samples from Separate Residences: A cross-sectional sample of homes with collected dust samples of approximately equal detection sensitivity was desired. Given the drop in detection frequency with lower mass extracted, and high number of samples in the highest mass bin, a minimum 0.15 g aliquot of sieved mass was considered optimal for the analytical method. We created a dataset where samples from each of the separately sampled homes with sufficient collected mass to have allowed at least a 0.15 g aliquot to have been taken, were represented. Specifically, for those participants who had repeat samples from the same home, (i.e., between the three visits, 96 had not moved at all with three samples collected from the same house, and 64 had moved only once with two samples collected from the same house), we selected one sample from the two or three samples from the same house. That is, we selected the one sample either in the highest sieved mass aliquot category (i.e., >0.15 g) or if all samples were in that aliquot category, we randomly selected one sample. From those participants who had moved only once (n=64), we then added to the dataset the samples from the second home in which they had lived. If they had moved a second time (n=8), we added the samples from all three homes in which the participant had lived resulting in 248 samples (i.e., 96+64+64+(8*3)). From these 248 samples, results from seven homes were eliminated due to low sample mass category (<0.15 g) (n=241). We then eliminated two more samples from homes which were outside of the SV (n=239). As described in the main article, we further culled the data set to include only results from batches with average SRS recovery >75%.

Selection of Independent Variables: Multivariate regression may only support a number of variables equal to about 10-20% of the sample size (9). As PUR and weather variables were included in our models, we limited the analysis to the identified 11 variables. All of the selected information was collected at the sample collection home visit. In addition, collapsing responses into dichotomous variables assumes no monotonic increase between categories. Notably, the following decisions were made:

Farmworkers Living in the Home: We identified whether any members of the household worked in agriculture or did field work. In addition, the mothers were asked in the clinic interview if they handled or applied pesticides on the job. Only one participant did. Because occupational pesticide task information was not obtained from all residents in the house, we could not include a variable that stated whether or not any household member worked with or applied pesticides.

Storing of Work Shoes or Clothes: At the home-visit, participants were asked where they stored work shoes or clothes. If these items were kept anywhere inside the residence, except the garage, the items were considered stored inside the home.

Vacuuming/Cleaning: We did not include vacuuming/cleaning variables because all but three participants reported having cleaned their house in the past week.

Air Conditioner in Home: Having an air conditioner in the home was defined as having a central air conditioner, or having a window air conditioner or an evaporative cooler, also known as a “swamp cooler,” in any room. Of the 8% who had air conditioning, most had central air conditioning.

Living near a golf course: A very small percentage (4%) of our participants lived within a quarter of a mile of a golf course (km). Because of this small percentage, this variable was not included in our analysis.

Regression Analysis: All statistical analysis was done with SAS (version 9; SAS Institute Inc., Cary, NC). Tobit regression analysis was done with the PROC QLIM command. Results of bivariate regression analysis of the independent predictor variables with dust concentrations are given in Table S.6.

Mapping of Pesticide Use and Dust Concentrations:

To understand the geographical distribution of pesticide use in the Valley, we mapped the annual average applications of the five pesticides chlorpyrifos, diazinon, DCPA, iprodione, and permethrin for the study years 1999-2001 by PLSS section. We then overlaid the average pesticide dust concentrations for homes in each section with and without farmworkers (Figures S.1-S.5).

These maps generally confirmed the findings of the regression analysis. That is, those pesticides showing statistically significant associations between agricultural applications and dust concentrations, (i.e., chlorpyrifos, DCPA, and iprodione,) also showed similar geographical patterns of applications and dust concentrations in all homes (Figures S.1a-S.5a) and in homes without farmworkers (Figures S.1b-S.5b). Specifically:

- Chlorpyrifos dust concentrations were apparently higher in homes in the *central* portion of the SV, where agricultural applications of chlorpyrifos were higher, and were lower in the area of the city of Salinas (Figure S.1).
- For DCPA, dust concentrations and applications were higher in the *south* of the SV (Figure S.3).
- For iprodione, dust concentrations and applications were higher in the *north* of the SV (Figure S.4).
- For diazinon (Figure S.2) and permethrin (Figure S.5), which showed no statistically significant relationship between agricultural applications and dust levels in regression analysis, dust concentrations reveal no particular geographical pattern. However, agricultural applications of both, particularly diazinon, are higher in the north of the SV.

Table S.1: Average Annual kg of Pesticides Used Agriculturally in the Salinas Valley in 1999-2002^a

<u>Pesticide</u>	<u>Kg applied</u>
methyl-bromide	1,028,656
chloropicrin	707,090
sulfur	642,807
1,3-dichloropropene	154,233
maneb	143,387
fosetyl-al	77,917
metam-sodium	64,890
diazinon	57,223
malathion	41,029
chlorthal-dimethyl	34,811
sodium-tetrathiocarbonate	33,777
acephate	33,625
mineral oil	31,504
lime-sulfur	28,080
oxydemeton-methyl	27,673
methomyl	26,919
chlorpyrifos	25,913
captan	24,461
<u>dicloran</u>	23,130

^a A total of 472 different pesticides were applied in the Salinas Valley in the years 1999-2002. Only those pesticides with use greater than 23,130 kg (50,000 pounds) per year are listed.

Table S.2: Pesticide Use in Salinas Valley and Environmental Fate Physicochemical Parameters of Dust Analytes^a

	CAS number	Agricultural Pesticide Use in SV: (average kg/yr: 1999-2001)	Other non- agricultural pesticide use in SV ^b (average kg/yr: 1999-2001)	Molecular Weight (g/Mol)	Vapor Pressure (mmHg @25°C)	Water Solubility (mg/L @25°C)	Log K _{ow} (octanol / water)
<u>OP Insecticides</u>							
diazinon	333-41-5	57,223	877	304	9*10 ⁻⁵	60 (@22°C)	3.3
malathion	121-75-5	41,029	137	330	1.8*10 ⁻⁴	130	2.7
acephate	30560-19-1	33,625	318	278	1.7*10 ⁻⁶	818,000 (@20°C)	-0.85*
oxydemeton	301-12-2	27,673	0	246	2.9*10 ⁻⁵ (@20°C)	Miscible	-0.74*
chlorpyrifos	2921-88-2	25,913	745	351	2.0*10 ⁻⁵	1.2	5.0
bensulide	741-58-2	14,417	22	398	8.0*10 ⁻⁷	5.6	4.1
methidathion	950-37-8	6,388	3	302	3.4*10 ⁻⁶	187 (@20°C)	2.2
fonofos	944-22-9	1,564	0	246	3.4*10 ⁻⁴	13	3.9
fenamiphos	22224-92-6	1,357	35	303	1.0*10 ⁻⁶	330 (@20°C)	3.2
phosmet	732-11-6	1,183	8	317	4.9*10 ⁻⁷	20	3.0
methamidophos	10265-92-6	604	0	141	3.5*10 ⁻⁵	Miscible (@20°C)	-0.8
azinphosmethyl	86-50-0	203	0	317	1.6*10 ⁻⁶ (@20°C)	29	2.8
<u>Other Analytes</u>							
DCPA	1861-32-1	34,811	12	332	2.5*10 ⁻⁶	0.5	4.4*
methomyl	16752-77-5	26,919	3	162	5.4*10 ⁻⁶	58,000	0.57
iprodione	36734-19-7	22,443	103	330	3.75*10 ⁻⁹	13.9	3.1
dimethoate	60-51-5	16,685	0	229	1.9*10 ⁻⁵	23800	0.78*
permethrin	52645-53-1	11,941	362	391	2.2*10 ⁻⁸	0.006	6.1
vinclozoline	50471-44-8	6,653	16	286	1.2*10 ⁻⁷ (@20°C)	2.6	3.1
DDE	72-55-9	Not used	Not used	318	6.0*10 ⁻⁶	0.0012	6.9
DDT	50-29-3	Not used	Not used	355	1.6*10 ⁻⁷ (@20°C)	0.04	6.9

(a) Vapor pressure obtained HSDB (10) water solubility and log K_{ow} obtained from ARS (11) or, if not available there (*) are from HSDB.

(b) To estimate non agricultural pesticide use in the SV, the non-agricultural uses for Monterey, Santa Cruz, and San Benito Counties were weighted by the percentage of land mass in the county in the SV (83, 13, and 3%, respectively) and summed.

Table S.3: QC Results for Spiked Dust and Duplicate Dust Analyses

Analysis method/ Class	Analyte	Recovery from Spiked Dust, Ave % \pm stdev		Duplicate Dust Samples (n=14)	
GC/MS		100 ng/g (n=13)	1000 ng/g (n=11)	Detects, n	Average RPD
Non-polar	chlorthal-dimethyl	95 \pm 23	103 \pm 25	14	28
	p,p'-DDE	113 \pm 57 ^a	105 \pm 13	8	13
	p,p'-DDT	NC ^b	127 \pm 63	7	24
Moderately polar	azinthos methyl	NC ^c	124 \pm 80	0	--
	chlorpyrifos	106 \pm 15	111 \pm 15	13	18
	diazinon	105 \pm 26	102 \pm 12	13	32
	fonofos	91 \pm 22	97 \pm 15	0	--
	iprodione	108 \pm 29	94 \pm 11	8	18
	malathion	151 \pm 38	112 \pm 13	4	50
	methidathion	131 \pm 63	111 \pm 13	0	--
	cis-Permethrin	NC ^d	100 \pm 35	14	20
	trans-Permethrin	NC ^e	101 \pm 33	14	14
	phosmet	109 \pm 21	105 \pm 19	0	--
	vinclozoline	116 \pm 22	107 \pm 12	0	--
	acephate	NC ^f	87 \pm 28	0	--
Polar	chlorpyrifos oxon	109 \pm 93	87 \pm 18	0	--
	dimethoate	NC ^f	130 \pm 25	0	--
LC/MS/MS		50 ng/g (n=8)	500 ng/g (n=10)		
Moderately polar	bensulide	97 \pm 57	91 \pm 41	13	67
	fenamiphos	96 \pm 53	103 \pm 28	1	200
Polar	methamidophos	50 \pm 21	47 \pm 22	4	115
	methomyl	96 \pm 22 ^g	49 \pm 16	11	47
	oxydemeton methyl	100 \pm 41	50 \pm 15	22	54

- a) Native dust level is high relative to spike level (0.5X spike level); thus high variability expected
b) No calculation; native dust level is high relative to spike level (3X spike level); thus cannot calculate recovery
c) No calculation; interference compound in dust is high relative to spike level; thus cannot calculate recovery
d) No calculation; native dust level is high relative to spike level (15X spike level); thus cannot calculate recovery
e) No calculation; native dust level is high relative to spike level (18X spike level); thus cannot calculate recovery
f) Spike level was close to detection limit; thus cannot calculate recovery
g) Recovery in 3 samples; ND in other 5 samples; 70% recovery would have been below the detection limit

Table S.4: Average and distribution of SRS recoveries in 55 QC samples and 504 field samples by GC/MS

SRS	Polarity	% SRS Recovery, ave \pm stdev		Distribution of SRS recoveries in field samples		
		QC samples (n=55)	Field samples (n=504)	>125%	75-125%	<75%
¹³ C ₁₂ -p,p'-DDE	non-polar	13 \pm 7	11 \pm 6	NA ^a	NA	NA
¹³ C ₆ -cis-permethrin	slightly polar	81 \pm 19	79 \pm 22	0.8%	63%	37%
¹³ C ₆ -trans-permethrin	slightly polar	83 \pm 20	79 \pm 22	0.8%	63%	37%
Fenchlorphos	moderately polar	82 \pm 16	79 \pm 19	0.4%	77%	23%
¹³ C ₁ -DEAA	polar	79 \pm 19	83 \pm 20	1.8%	72%	26%

NA= not applicable; planned loss of SRS to remove potential interferences

Table S.5: QLs, Detection Frequency, and Concentration Range by Different Ranges of Dust Mass Extracted

GC/MS Analysis	QL (ng/g) by mass extracted,			Detection frequency (%) by mass extracted				Concentration (ng/g) by dust mass,		
	0.15-0.50 g	0.05-0.14 g	0.01-0.04 g	Overall (n=504)	0.50-0.15 g (n=432)	0.14-0.05 g (n=50)	0.04-0.01 g (n=22)	0.15-0.50 g	0.05-0.14 g	0.01-0.04 g
Non-polar										
DCPA	2	7	20	92	95	88	41	0.3-1401	7-256	32-147
p,p'-DDE	2	7	20	35	40	10	0	5-305	9-125	ND
p,p'-DDT	10	30	100	39	43	22	0	30-998	11-1849	ND
Moderately polar										
azinphos methyl	200	700	2000	4.6	4.9	4.0	0	19-8555	138-414	ND
chlorpyrifos	2	7	20	82	86	68	4.5	0.3-8678	6-9808	698
diazinon	2	7	20	82	87	64	23	2-7175	16-1419	80-1700
fonofos	4	12	40	0.4	0.2	2.0	0	0.5-44	43	ND
iprodione	10	30	100	42	46	20	14	6-8808	39-1298	69-784
malathion	10	30	100	11	12	4.0	9.1	32-1063	305-2984	386-1149
methidathion	10	30	100	1.6	1.9	0.0	0	28-721	ND	ND
cis-permethrin	5	15	50	96	98	96	59	10-49,795	67-4205	131-5423
trans-permethrin	5	15	50	97	99	98	59	15-221,866	12-5671	47-3225
phosmet	10	30	100	7.5	7.9	6.0	4.5	17-5941	1-7786	2469-2469
vinclozoline	10	30	100	2.8	3.2	0	0.0	16-502	ND	ND
Polar										
acephate	40	130	400	2.6	3.0	0	0.0	15-9189	ND	ND
chlorpyrifos oxon	20	65	200	0.2	0.2	0	0.0	491-491	ND	ND
dimethoate	50	165	500	0.4	0.2	2.0	0.0	45-45	789	ND
LC/MS/MS				(427)	(412)	(15)	none			
Moderately Polar										
bensulide	30	90	300	22	22	13	--	1-537	22-174	--
fenamiphos	5	15	50	3.3	3.4	0	--	1-459	ND	--
Polar										
methamidophos	5	15	50	11	11	6.7	--	3-115	5	--
methomyl	60	180	600	32	31	53	--	16-518	19-210	--
oxydemeton	5	15	50	62	63	40	--	0.3-100	11-58	--

Table S.6: Bivariate Regression Results: Proportional Change in Dust Concentrations for Unit Change in Predictor Variables^a

<u>Parameter</u>	<u>chlorpyrifos</u> (n=197)		<u>Diazinon</u> (n=197)		<u>DCPA</u> (n=181)		<u>iprodione</u> (n=181)		<u>trans-pemethrin</u> (n=181)		<u>cis-pemethrin</u> (n=177)	
	Mean	PC	Mean	PC	Mean	PC	Mean	PC	Mean	PC	Mean	PC
agricultural pesticide use ^b prior to sample collection												
days 1-3	0.10	-0.03	0.55	0.00	0.27	0.04	0.27	-0.16	0.13	-0.57	0.13	-0.51
month (days 4-33)	0.27	0.27 *	1.5	0.00	1.04	0.11 ***	1.05	0.21 ***	0.45	0.03	0.45	0.00
season (days 34-133)	0.46	0.05	1.7	-0.01	1.14	0.17 ***	1.03	0.26 ***	0.45	-0.08	0.45	-0.59
rainfall (mm per day) prior to sample collection:												
days 1-3	0.59	-0.01		-0.04 **	0.59	-0.03 **		0.00		-0.01	0.59	-0.01
month (days 4-33)	0.85	0.04		-0.01	0.89	-0.06 ***		-0.06		0.01	0.89	0.01
season (days 34-133)	0.91	0.02		0.01	0.92	-0.06 *		-0.10		0.04	0.92	0.03
temperature (mean daily °C) prior to sample collection:												
days 1-3	21	-0.01		-0.01	21	0.04 ***		0.03		-0.02	21	-0.02
month (days 4-33)	21	-0.01		0.00	21	0.06 ***		0.05		-0.01	21	-0.01
season (days 34-133)	21	0.00		0.01	21	0.07 ***		0.06		-0.01	21	0.00
Home Visit Information (Y/N)	<u>%yes</u>				<u>%yes</u>						<u>%yes</u>	
living within 60 m of a field	9	1.00		-0.76	8	1.13 *		-0.10		-0.12	8	-0.22
farmworkers in home	76	0.28		1.32 **	76	0.95 ***		5.79 ***		0.13	77	0.13
work shoes stored in home	22	1.52 **		1.29 **	23	0.59 *		0.59		0.99 *	23	0.76 *
work clothes stored in home	52	0.77 *		0.90 **	51	0.44 *		2.74 **		0.79 *	53	0.70 *
chlorpyrifos product in home	3	-0.44		NA		NA		NA		NA		NA
diazinon product in home	2	NA		11.99 **		NA		NA		NA		NA
pyrethrin/pyrethroid in home		NA		NA		NA		NA	27	0.36	27	0.22
home pesticide application ^c	8	0.36		-0.28	7	-0.43		-0.35		-0.23	7	-0.24
any pets	23	0.01		-0.02	24	0.38		-0.42		-0.30	24	-0.31
housing density > 2 per room	25	0.08		1.02 **	24	0.36		-0.04		0.25	25	0.18
home less clean	72	0.54		-0.12	70	0.76 **		0.48		0.21	71	0.11
air conditioner in home	8	-0.81 **		-0.53	6	-0.17		-0.63		1.75 *	6	1.12
furniture sample collected	14	-0.32		-0.19	14	-0.03		0.03		-0.20	15	-0.26

- a) All dust levels are log transformed. PC is proportional change and is equal to $((10^b) - 1.0)$, where b is the beta regression coefficient; * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$. All home visit information coded 1=yes, 0=no.
- b) Kg/day of agricultural pesticide use near home (23 km²) prior to sample collection.
- c) Professional pesticide application at home within the past 6 months prior to sample collection.

Figure S.1a and S.1b: Chlorpyrifos use and dust concentrations in the Salinas Valley

Chlorpyrifos

Chlorpyrifos Levels in House Dust and
Chlorpyrifos Agricultural Applications in
Salinas Valley, California
1999 - 2002

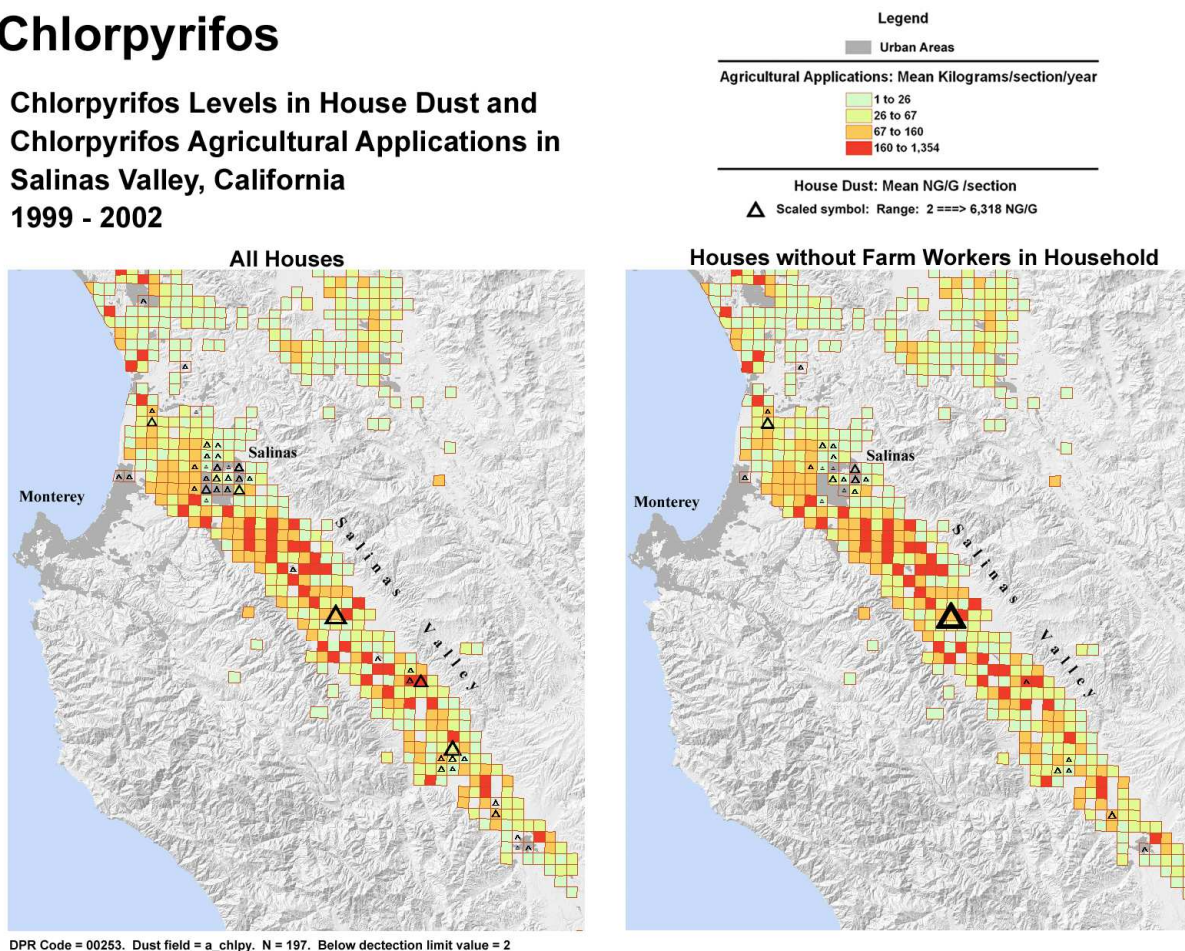
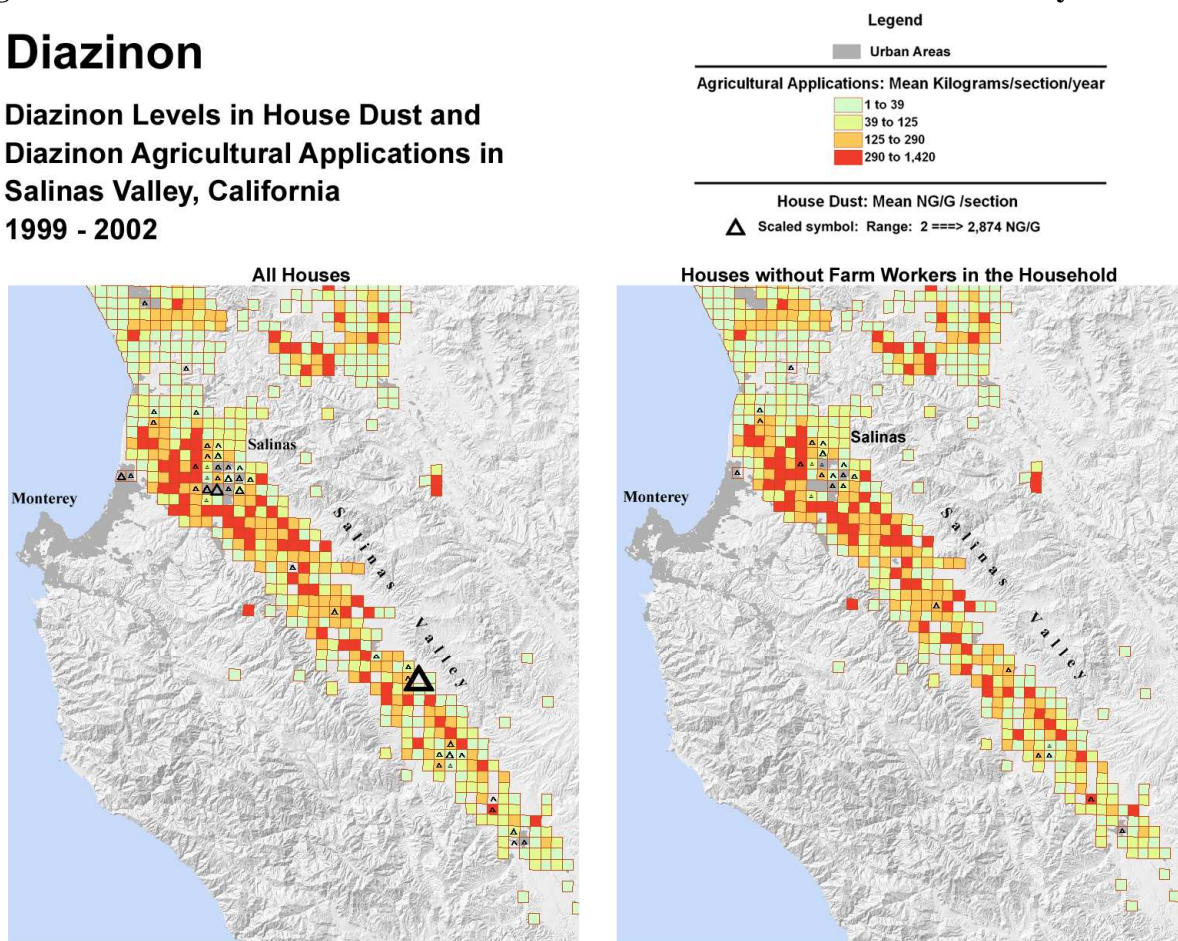


Figure S.2a and S.2b: Diazinon use and dust concentrations in the Salinas Valley

Diazinon

Diazinon Levels in House Dust and Diazinon Agricultural Applications in Salinas Valley, California 1999 - 2002



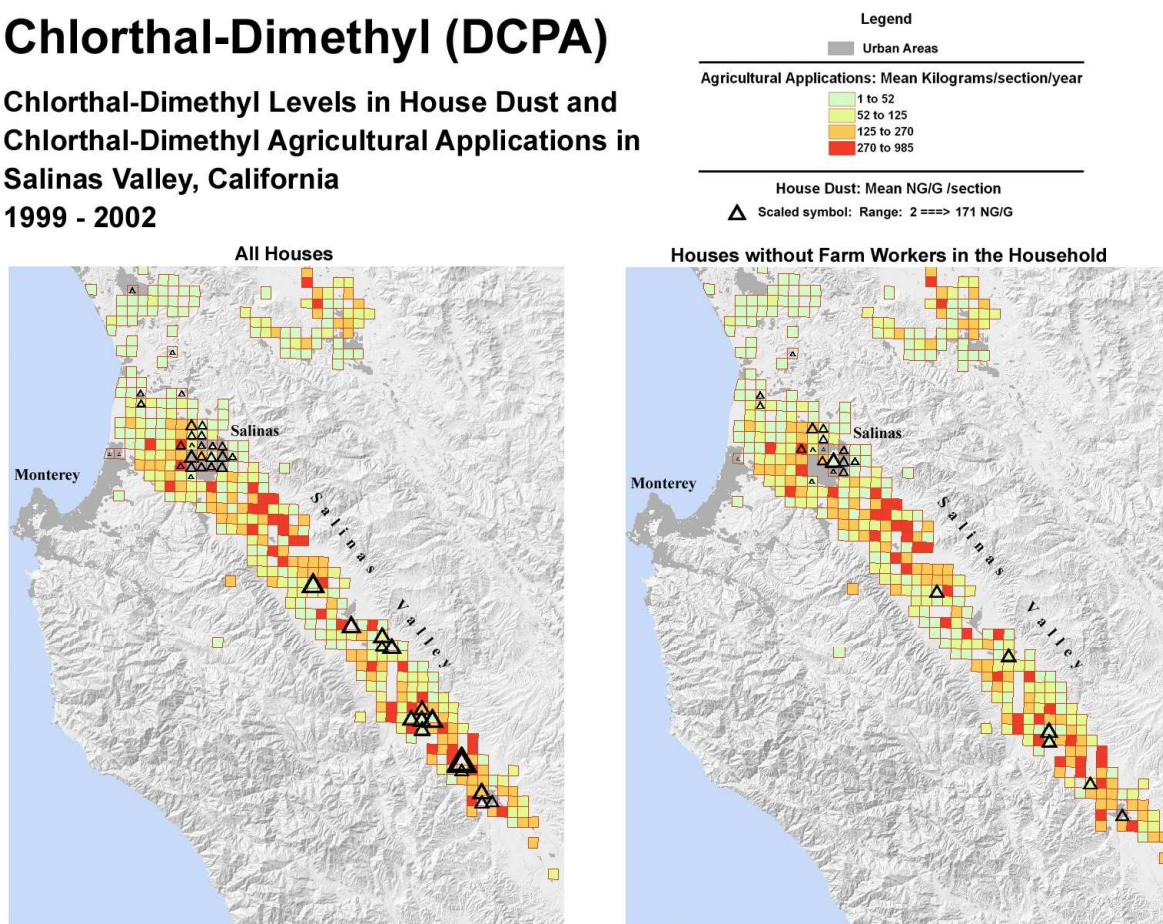
DPR Code = 00198. Dust Field = a_diaz. N = 197. Below detection limit value = 2.

Figure S.3a and S.3b: DCPA use and dust concentrations in the Salinas Valley.

Chlorthal-Dimethyl (DCPA)

Chlorthal-Dimethyl Levels in House Dust and
Chlorthal-Dimethyl Agricultural Applications in
Salinas Valley, California

1999 - 2002

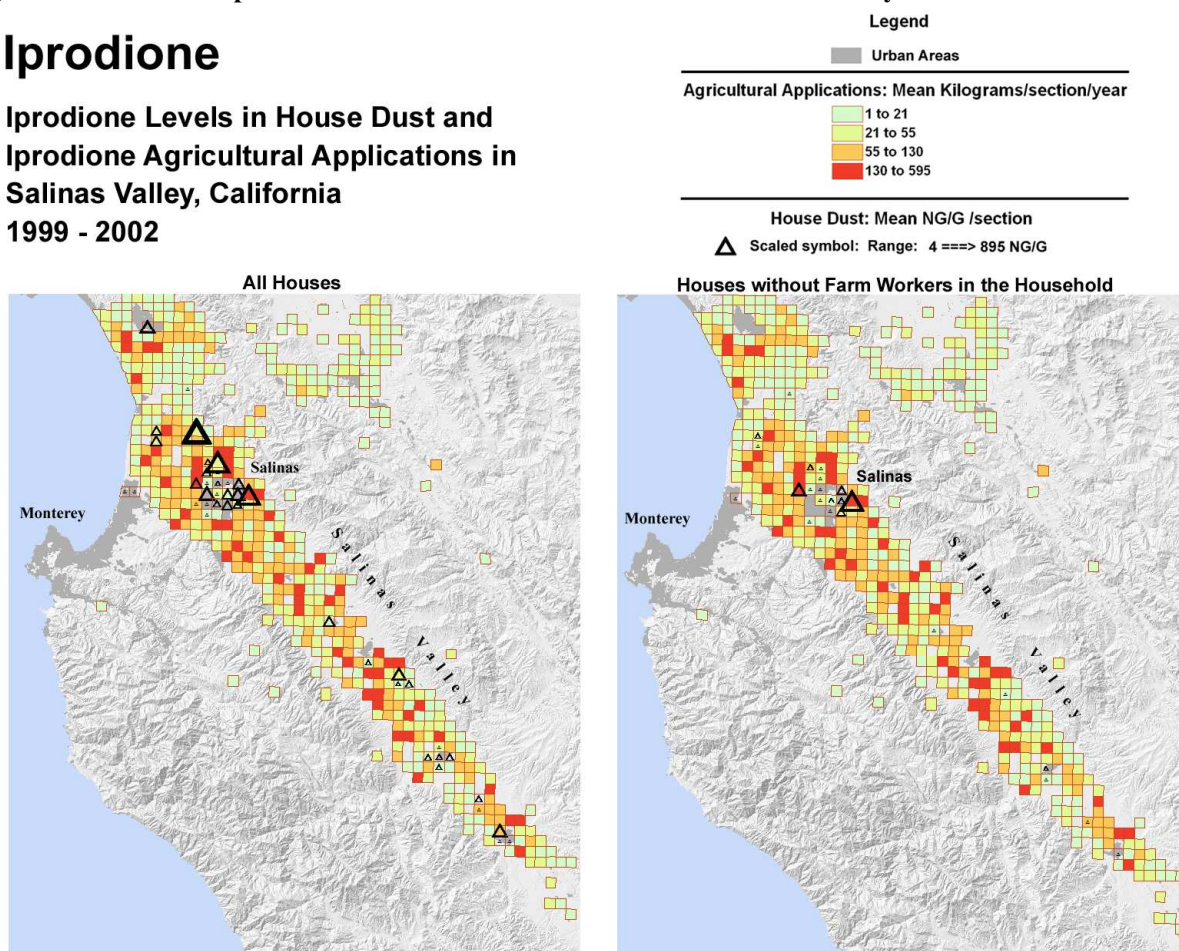


DPR Code = 00179. Dust field = a_dacth. N = 181. Below detection limit value = 2.

Figure S.4a and S.4b: Iprodione use and dust concentrations in the Salinas Valley

Iprodione

Iprodione Levels in House Dust and Iprodione Agricultural Applications in Salinas Valley, California 1999 - 2002

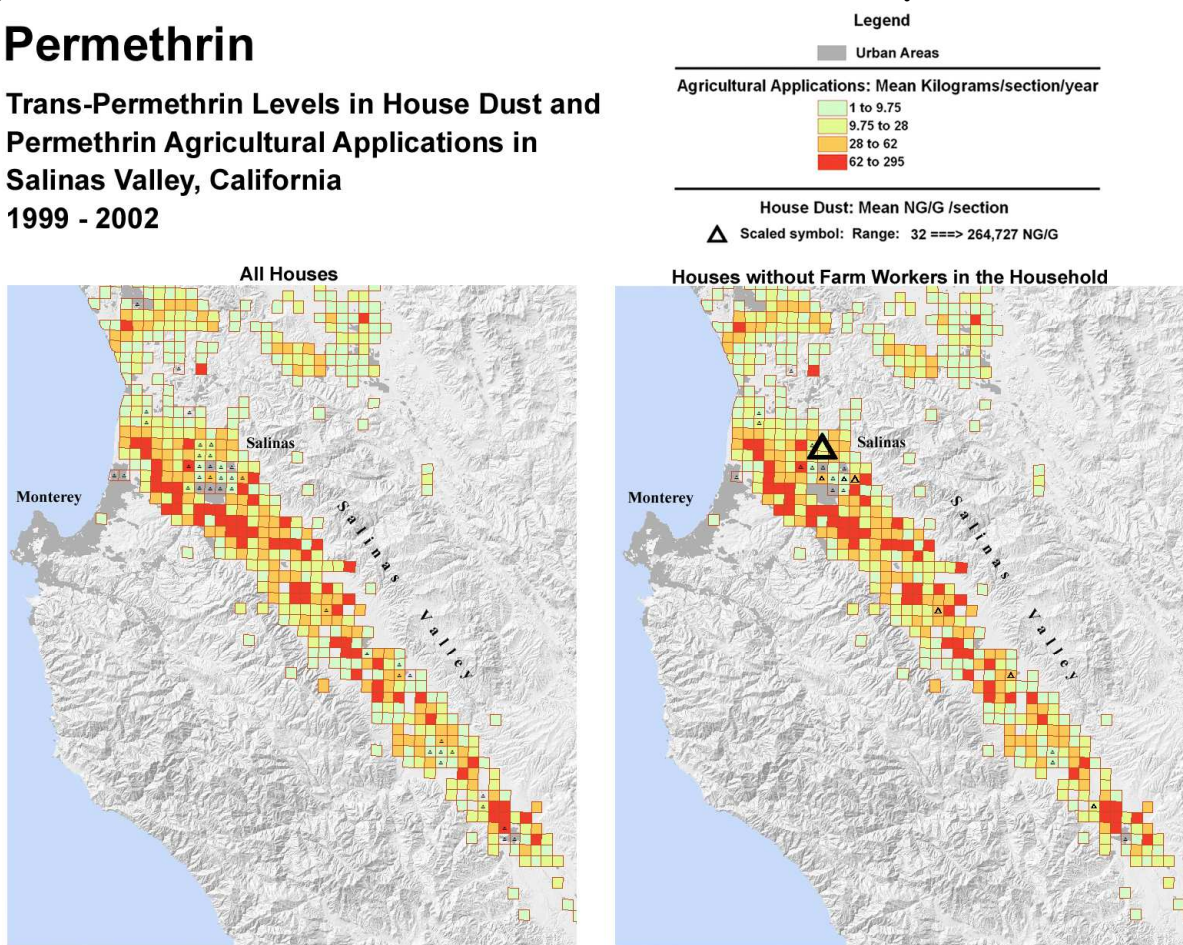


DPR Code = 02081. Dust Field = a_iprod. N = 181. Below detection limit value = 4.

Figure S.5a and S.5b: Permethrin use and dust concentrations in the Salinas Valley.

Permethrin

Trans-Permethrin Levels in House Dust and
Permethrin Agricultural Applications in
Salinas Valley, California
1999 - 2002



DPR Code = 02008. Dust Field = a_permt. N = 181. Below detection limit value = 10.

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