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Determinants of pesticide concentrations in silicone wristbands worn by Latina adolescent girls in a California farmworker community: The COSECHA Youth Participatory Action Study

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Abstract

Personal exposure to pesticides has not been well characterized, especially among adolescents. We used silicone wristbands to assess pesticide exposure in 14 to 16 year old Latina girls (N = 97) living in the agricultural Salinas Valley, California, USA and enrolled in the COSECHA (CHAMACOS of Salinas Examining Chemicals in Homes and Agriculture) Study, a youth participatory action study in an agricultural region of California. We determined pesticide concentrations (ng/g/day) in silicone wristbands worn for one week using gas chromatography electron capture detection and employed gas chromatography mass spectrometry to determine the presence or absence of over 1500 chemicals. Predictors of pesticide detections and concentrations were identified using logistic regression, Wilcoxon rank sum tests, and Tobit regression models. The most frequently detected pesticides in wristbands were fipronil sulfide (87%), cypermethrin (56%), dichlorodiphenyldichloroethylene (DDE) (56%), dacthal (53%), and trans-permethrin (52%). Living within 100 m of active agricultural fields, having carpeting in the home, and having an exterminator treat the home in the past six months were associated with higher odds of detecting certain pesticides. Permethrin concentrations were lower for participants who cleaned their homes daily (GM: 1.9 vs. 6.8 ng/g/day, $p= 0.01$). In multivariable regression models, participants with doormats in the entryway of their home had lower concentrations ($p<0.05$) of cypermethrin (87%), permethrin (99%), fipronil sulfide (69%) and DDE (75%). The results suggest that both nearby agricultural pesticide use and individual behaviors are associated with pesticide exposures.

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Conflict of Interest
Kim Anderson, an author of this research, discloses a financial interest in MyExposome, which is marketing products related to the research being reported. The terms of this arrangement have been reviewed and approved by Oregon State University in accordance with its policy on research conflicts of interest.

Keywords

Pesticides; exposure assessment; personal monitoring; silicone wristbands; youth participatory action research

1. Introduction

As the leading agricultural state in the United States, California uses more than 185 million pounds of pesticides each year, including several pesticides that have been identified as probable or possible carcinogens, endocrine disruptors, or reproductive and developmental toxicants.[1] Exposures to some pesticides has been associated with earlier puberty, menstrual cycle irregularities, impaired fertility, disruption of hormonal function and increased breast cancer risk.[2-4] Puberty, a time of rapid reproductive development, breast tissue proliferation, and brain growth, may be a critical window for long-term health effects of pesticide exposure. Thus, young girls in agricultural communities may be particularly vulnerable to pesticides in their environment. To date, few studies have examined the extent to which adolescent girls living in agricultural communities are exposed to pesticides from nearby agricultural use.

Research has shown that residents of agricultural communities have higher levels of certain pesticide metabolites in their urine[5-6] as well as higher levels of some agricultural pesticide in their house dust[7-10] compared with non-agricultural populations. However, there is still much that is not known about pesticide exposure to residents of rural areas, particularly adolescents. Measurement of pesticide biomarkers in urine or blood is considered the gold standard for exposure assessment, but the vast majority of pesticides currently used in agriculture lack established biomarkers. Similarly, household dust has been used as a matrix for estimating exposure, but is only a surrogate for potential exposures in the home and misses exposure that occurs elsewhere.

Silicone wristbands, which can capture hundreds of chemicals in a simple and nonintrusive manner, offer a promising new technology for passive personal monitoring of pesticides among residents of agricultural communities.[11] As a passive sampling device, silicone wristbands non- selectively sequester bioavailable volatile and semi-volatile organic compounds via diffusion, similar to a phospholipid membrane.[11] The bioavailable fraction is significant for inhalation and dermal exposure routes in humans,[12-15] and these exposure routes can significantly increase the risk of adverse health outcomes.[13-14] Silicone wristbands have been used to assess exposure for VOCs in asphalt workers,[11] flame retardants for preschool age children,[12] pesticides for farm workers in Africa, and pesticides for people living in both agricultural and urban communities in Peru. Because silicone wristbands provide the capacity to measure personal exposures to pesticides both at home and away, they present an opportunity to examine predictors and correlates of exposure.

The Salinas Valley in California's Monterey County is one of the most concentrated and productive agricultural regions of the state, with annual revenues from agriculture exceeding four billion dollars per year.[16] Agricultural pesticide use in this area is high with approximately 9 million pounds applied per year.[1] The Center for the Health Assessment of

Mothers and Children of Salinas (CHAMACOS) is a community-based participatory research partnership investigating pesticide exposure and health effects in this community, through a longstanding longitudinal birth cohort study, pesticide exposure studies, and youth participatory action research. CHAMACOS serves the farmworker community of the Salinas Valley, a population comprised predominantly of low-income, Spanish-speaking immigrants from Mexico and their families. We have previously assessed pesticide exposure to residents of this community via metabolites of organophosphate pesticides in urine[17] and concentrations of several pesticides in dust [18-19]. Among pregnant women in the CHAMACOS Cohort Study, median urinary concentrations of metabolites of organophosphate pesticides were approximately 40% higher than those in a representative sample of U.S. women in the National Health and Nutrition Examination Survey (NHANES).[17] In house dust samples, we observed higher concentrations of several pesticides in homes with recent nearby agricultural pesticide use, farmworkers living in the home, and farmworkers storing their work clothes or shoes in the home.[18-19] Although these studies suggest higher pesticide exposure to participants living with farmworkers and near agricultural fields, personal monitoring had not been performed in this community.

In the present study, we report findings from personal monitoring using silicone wristbands worn by adolescent girls living in the Salinas Valley. Study participants were part of a youth-led participatory action project that was embedded in the CHAMACOS Longitudinal Cohort Study. The goal of this sub-study, known as the CHAMACOS of Salinas Examining Chemicals in Homes and Agriculture (COSECHA) Study, was to determine predictors of pesticides detected on silicone wristbands worn by adolescents girls living in the Salinas Valley, while also engaging and empowering local youth in peer-to-peer research, education, and advocacy.

2. Materials and methods

2.1 Youth Participatory Action Research Design

The COSECHA study was developed as a youth participatory action project by the CHAMACOS Youth Council, a group of local high school students that serves as both a youth advisory board to the CHAMACOS Center and a venue for engaging local youth in environmental health awareness and education. The Youth Council identified pesticide exposure to adolescents as an important issue in their community and met regularly with researchers to learn how to design and implement the study. Ten Youth Council members were hired as summer research assistants and conducted for all aspects of data collection. The youth, most entering their junior year of high school, were thoroughly trained for at least 6 months prior to entering the field. In preparation for data collection, they participated in home visit simulations, piloted equipment in various settings, and learned about environmental health research methods. The study was designed as a peer-to-peer project, with youth researchers collecting data from and returning study findings to other adolescents in their community.

2.2 Study population.

Participants in the study were 14-16 year old girls living in the Salinas Valley and participating in the CHAMACOS longitudinal cohort study. Details about recruitment of CHAMACOS cohort participants have been published elsewhere.[20] Briefly, families with children born between 2000 and 2002 were recruited from health clinics, social service providers, and by word of mouth either during the mother's pregnancy or when the child was 9 years old. Families were eligible if, at the time of the pregnancy, the mother was over 18 years of age, spoke English or Spanish, received low-income state health insurance (Medi-Cal), and had planned to deliver at a local hospital. During the summer of 2016, a subset of 100 girls was selected from among the 609 boys and girls participating in the CHAMACOS cohort at age 14. Girls were eligible for the COSECHA sub-study if they lived in Monterey or San Benito Counties and had a cell phone to receive daily text reminders.

Wristbands were deployed to all 100 participants. One study participant lost her wristband and, for two participants, wristband pesticide concentrations could not be quantified due to matrix interference, leaving a final sample size of 97 girls. Research protocols were approved by the University of California, Berkeley, Committee for the Protection of Human Subjects. We obtained written informed consent from mothers and written assent from teenaged daughters.

2.2 Data collection.

All data collection was conducted by trained Youth Research Assistants (YRAs), with supervision from an adult staff member, at two home visits spaced approximately one week apart. Data collection was conducted between June 3 and Aug 4, 2016, during the summer agricultural season when pesticide applications were expected to be high. At the first home visit, the participant removed the wristband from its sealed storage bag and placed it on her wrist. We asked participants to wear the wristband continually for seven days, even when showering or sleeping. At the second home visit, the participant returned the wristband to its sealed bag and it was placed in a −20°C freezer until shipment to Oregon State University for analysis.

At the first home visit, YRAs conducted a home survey to document housing characteristics (e.g. presence of a doormat, carpeting in the home) and a home pesticide inventory to record all pesticide products in the home (including the product name and United States Environmental Protection Agency (USEPA) registration number). An adult staff member verified the active ingredients of pesticides found in the home based on the USEPA registration numbers recorded by the YRAs. For each home, the YRAs printed out a Google Earth map in advance of the visit which showed the location of all fields within 1/4 mile of the residence. The YRA then visited each nearby field, confirmed its location on the map, visually identified which crops were growing there, and marked this information on the map. If the crops were too young to identify or not recognizable to the YRA, the YRA and an adult staff member revisited the area 1-3 weeks later to confirm the crops.

At the second home visit, YRAs administered a structured questionnaire to the adolescent participants about factors potentially related to pesticide exposures, including the number of agricultural workers living in her home, insecticide use in the home during the previous six months, treatment by a professional exterminator during the previous six months, whether she observed aerial spraying near her home during the sampling period, frequency of home cleaning, and how often the windows were open during the sampling period. Participants received daily text messages during the week reminding them not to remove the wristband and providing a number to call if they had questions or problems.

2.3 Wristband Passive Sampling.

The silicone wristbands (https://24hourwristbands.com, Houston, TX; mass: 4.1 ± 0.1 g; width: 1.3 cm) were purchased online. The conditioning, post-deployment cleaning, and extraction of the wristbands were performed as described previously.[21-22] Briefly, the wristbands were conditioned at 300°C for 180 minutes and under vacuum at 0.1 Torr and stored in airtight Teflon bags prior to deployment. For post-deployment cleaning, the wristbands were rinsed with 18 MΩ cm water and isopropanol to remove particulate matter. The wristbands were subsequently extracted in two 100 mL volumes of ethyl acetate, which combined and quantitatively reduced to 1 mL. A 200 μL aliquot of extract underwent solid-phase extraction (SPE) on a C18 silica column with acetonitrile, followed by a solvent exchange to iso-octane. All solvents were Optima-grade or equivalent (Fisher Scientific, Pittsburgh, PA). All analytical grade standards purchased from Accustandard (New Haven, CT) were of 95% or higher purity. The pesticide extraction surrogates were tetrachloro-meta-xylene (TCMX) and decachlorobiphenyl. The internal standard was 4,4'-dibromooctafluorobiphenyl.

2.4 Chemical Analysis in Wristbands.

Two analytical methods were performed on each wristband. The first method used gas chromatography (GC) dual micro-electron capture detection (μECD) and yielded quantitative time-weighted average concentrations of 72 pesticides. This method is based on the US EPA pesticide method 8081B, modified from Donald et al 2016,[23] and demonstrated in Vidi et al 2017[24]. An Agilent 6890N GC with dual 7683 μECD injectors was used with a DB-5MS and a DB-17MS column (each 30 m length, 0.25 mm diameter, 0.25 μm film thickness). The oven profile was set with an initial hold at 110°C for 0.5 minute, followed by a ramp to 150°C at 25°C/min, a second ramp to 229°C at 6°C/min, and a final ramp to 320°C at 20°C/min before holding at 320°C for 2.5 minutes. Donald et al 2016[23] described the confirmation process for identifying the target analytes and the determination of instrument limits of detection (LODs) over the course of three analytical days (n=7). Instrument LODs for the pesticides detected in >10% of wristbands using the quantitative method are listed in Table 1. Concentrations of the herbicide simazine could not be quantified due to interference from two fragrance compounds with similar molecular structures.

The second method screened for the presence or absence of over 1,500 chemicals, including 842 legacy and current-use pesticides. The screening method used an Agilent 6890N GC with a 5975B Mass Selective Detector and a DB-5MS column with retention times and automatic mass spectral deconvolution and identification software at an electron impact of 70eV. The spectra were compared against in-house and purchased libraries (e.g., NIST). The screening method identified compounds with a level of confidence as tentative candidates (level 3), not definitive identification (level 1).[25] A list of the pesticides determined to be present by the screening method is reported in Supplemental Information; the complete list of compounds has been previously reported. The sensitivity of the screening method to identify the presence of compounds varied based on sample background and the software's ability to deconvolute a total ion chromatogram, and identify compounds with a reference library spectral match above 70. Based on pesticides analyzed using both methods, the screening method was less sensitive for identifying the presence of compounds than the quantitative method was for detecting compounds. Only two of the pesticides analyzed by this method, piperonyl butoxide and DEET, were identified as being present in >10% of wristbands and are included in Table 1.

2.5 Quality Control.

Conditioned wristbands were tested for data quality assurance via GC-MS and sealed in polytetrafluoroethylene bags prior to shipment. Over 60% of analyzed samples were QC samples, including wristband conditioning verification samples (n=6), instrument blanks (n=104), an SPE blank (n=1), extraction reagent blanks (n=2), post-deployment cleaning blanks (n=2), trip blanks (n=2), and continuing calibration verifications (CCV, n=32). For the quantitative pesticide method, all target analytes in the blank QC samples were below limits of detection. The CCVs had an average of 84.9% of target pesticide analytes within ±20% of the expected value, with a range of 75-91% across all analyses. TCMX and decachlorobiphenyl were used as extraction surrogate compounds to assess for loss during laboratory processes. The average surrogate recoveries for the wristband samples were 55 ± 17% (median = 55%; range = 11- 138%) for TCMX and 52 ± 21% (median = 48%; range = 22 - 142%) for decachlorobiphenyl. If the target analyte was present in a wristband sample by 10X fold above all blank QC samples, then the analyte was included in the statistical analysis. In all remaining blank QC samples, all target analytes were not present using the screening method.

2.6 Data analysis.

The outcomes of interest were: 1) the presence/absence of detectable pesticides in the wristbands (for all pesticides detected in >10% of wristbands), and 2) time-weighted concentrations (ng/g/day) in wristbands (for all pesticides detected in >50% of wristbands). Time-weighted pesticide concentrations were calculated using fractional days defined by the starting and ending time/date that the participant wore the wristband. Most participants (n=92) wore the wristbands for seven days.

We investigated factors associated with the presence/absence of pesticides using logistic regression to calculate bivariate (unadjusted) odds ratios. We examined rank correlations among time-weighted concentrations of pesticides using non-parametric Spearman correlation coefficients because the pesticide concentrations were not normally distributed. Because of the relatively large number of values <LOD, we examined potential determinants of pesticide concentrations as continuous variables using the non-parametric Wilcoxon rank

sum test for unadjusted associations and Tobit regression for multivariable prediction models. For values below the LOD, we substituted the LOD/sqrt(2). We used natural log-transformed pesticide concentrations to reduce the influence of outliers and improve the fit of Tobit regression models.

Potential predictors of pesticide presence/absence or pesticide concentrations were selected *a priori* based on whether the pesticide in question was currently used only in agriculture, only in residential settings, in both agriculture and residences, or was a historical pesticide that was no longer used in California (see Table 1). These classifications were determined based on agricultural pesticide use in Monterey County reported in the 2016 California Pesticide Use Reporting (PUR) database.[1]

For agricultural pesticides, we examined associations with factors related to living near agriculture (i.e. living within 100 m of agricultural fields, living with agricultural workers, observing aerial pesticide spraying from the home, living in the city of Salinas vs. outside of Salinas in more rural areas (based on residential address), and leaving the windows open at night). We also looked at the relationships between agricultural pesticides and home characteristics (i.e. the presence of carpeting, having a doormat, frequency of housecleaning).For residential pesticides, we examined predictors related to the home characteristics listed above as well as factors related to home pesticide use (i.e. report of using pesticides in the home in the past 6 months, having the home treated by a professional exterminator in the past 6 months, having pets in the home, and having pesticide products in the home at the time of the home visit). We examined all predictors in analyses of pesticides with both agricultural and residential use. For analyses of historical use pesticides, we examined associations with home characteristics and with living within 100 m of agricultural fields, living with agricultural workers, and living in rural areas vs. the city of Salinas.

For the multivariate Tobit models, predictors were included if they were either significant or close to significant ($p<0.1$) in the bivariate analysis of pesticide concentrations (Table 4). We calculated the percentage change in wristband concentrations using the beta coefficients (β) from the Tobit regression models using the following equation.

$$Percent\ change\ = [exp(\beta) - 1] * 100$$

We evaluated model fit from the multivariable Tobit models (pseudo-R^2) using the sum of squares from the regression model (SS_{reg}) and the total sum of squares (SS_{tot}) based on the following equation.

$$Pseudo - R^2 = SS_{reg}/SS_{tot}$$

3. Results

Twenty-five pesticides were detected or present in 10% or more of the wristbands worn by study participants (Table 1). The most frequently detected agricultural pesticides, and their detections frequencies, were dacthal (52.6%), chlorpyrifos (36.1%) and dimethoate (13.5%). Frequently detected pesticides used in both agricultural and residential settings include cypermethrin (55.7%), *cis* and trans-permethrin (48.5 and 51.5%) and esfenvalerate (41.2%). The most commonly detected chemical overall was fipronil sulfide (86.6%), a breakdown product of fipronil, which is used as a flea treatment on pets, by professional exterminators for termite and other pest control, and in agriculture and turf products. We also detected several pesticides no longer used in California, including propachlor (53.6%) which was discontinued in 1998 and DDE (55.7%), a breakdown product of DDT, which was banned in 1972.

Supplemental Table S1 gives a complete list of the pesticides measured and their frequencies of detection or presence in wristbands. The number of pesticides detected in each wristband using the quantitative pesticide method ranged from 0 to 20 with an average of 8 pesticides detected per wristband (Supplemental Figure 1).

We observed weak to moderate correlations (r=0.2 - 0.4) between concentrations of DDE and cypermethrin, dacthal, and *cis* and trans-permethrin, and between propachlor, cypermethrin and dacthal in wristbands (Supplemental Table S2). The concentrations of *cis* and *trans*- permethrin were highly correlated (r=0.86, Table S2), therefore we summed the concentrations of the two isomers for further analyses. Distributions of measured pesticide concentrations (ng/g/day) for the most frequently detected pesticides (i.e. detected in >33% of wristbands) are shown in Table 2. The highest median (p50) concentrations measured in wristbands were for fipronil sulfide (12.9 ng/g/day), Σpermethrin (7.6 ng/g/day), cypermethrin (2.3 ng/g/day) and propachlor (2.3 ng/g/day). These same pesticides also had the highest concentrations at the upper end of the distribution. Chloroneb (p95=51.6 ng/g/day) and esfenvalerate (p95=20.3 ng/g/day) also had high concentrations at the tail of the distribution, but the highest concentration measured was for Σpermethrin (493.6 ng/g/day).

The unadjusted odds ratios for detections or presence of selected pesticides in wristbands by participant characteristics are presented in Table 3. (Unadjusted odds ratios for all pesticides detected or present in >10% of wristbands are shown in Supplemental Table S3.) Dacthal was the only pesticide used only in agriculture with statistically significant predictors of detection. The odds of detecting dacthal were 3.1 times greater if a participant lived within 100 m of an agricultural field (95% CI: 1.0-9.5; p <0.05), and 3.7 times higher if she had any carpeting in her home (95% CI: 1.4-10.0; p <0.05). The odds of dacthal detection were lower if participants lived in the city of Salinas rather than more rural areas of the Salinas Valley (OR=0.2; 95% CI: 0.1-0.4; p <0.01). We observed higher odds of detecting permethrin when home-use pesticide products were present in the home (OR=2.9; 95% CI: 1.3-6.6; p<0.05) and lower odds when participants had a doormat present at the entryway of the home (OR=0.1; 95% CI: 0.1-0.9; p<0.05). Unexpectedly, there were lower odds of detection for permethrin if participants reported having used pesticides in the home in the past 6 months (OR=0.1; 95% CI: 0.05-0.9; p<0.05).The odds of detecting esfenvalerate was

2.7 times higher (95% CI: 1.2-6.5; p<0.05) if a participant left her window open at least once at night during the week of the study. In the screening analysis, piperonyl butoxide, a synergist used in many pesticide formulations, had a lower odds of being present in wristbands when a doormat was used at the home (OR=0.2; 95% CI: 0-0.7; p<0.01), or the participant's home was cleaned daily (OR=0.3; 95% CI: 0.1-0.8; p<0.05). There were increased odds of presence of piperonyl butoxide if an exterminator had sprayed in or around the home in the last six months (OR=9.7; 95% CI: 2.1-45.8; p<0.01). In addition, having a professional exterminator spray in or around the home within the last six months increased the likelihood of detecting fipronil (OR=7.0; 95% CI: 1.4-35.7; p<0.05) (Table S3C). Chloroneb, a fungicide that was not used in the Salinas Valley in 2016, was eight times more likely to be detected in wristbands if there was carpet in the home (Table S3D).

When we investigated factors associated with concentrations of the more commonly detected pesticides in bivariate analyses (Table 4), we found that participants living within 100 m of an agricultural field, versus those who did not, had higher geometric mean concentrations of dacthal (1.0 vs. 0.3 ng/g/day), Σpermethrin (8.6 vs. 2.3 ng/g/day) and DDE (0.8 vs. 0.4 ng/g/day) in their wristbands. Participants living in Salinas had a lower geometric mean concentration of dacthal (0.2 ng/g/day) compared to those living in rural areas of the Salinas Valley (1.1 ng/g/day). We found higher geometric mean concentrations of Σpermethrin among participants with any carpet in the home (3.0 vs. 2.7 ng/g/day), pesticides stored in the house (6.9 vs. 1.2 ng/g/day), or using a professional exterminator within the last six months (22.7 vs. 2.4 ng/g/day). Participants with doormats (2.3 vs. 21.7 ng/g/day) and whose home was cleaned daily (1.9 vs. 6.8 ng/g/day) had lower geometric mean concentrations of Σpermethrin. We did not observe significant differences in any pesticide concentrations if there was an agricultural worker in the home. We also did not observe any significant differences in pesticide concentrations in wristbands if the participants had pets in their home or had windows open at night.

In multivariable Tobit regression models (Table 5), concentrations of dacthal were 93% lower (95% CI: −97%, −84%) among participants living in Salinas compared to those not living in Salinas and 311% higher (95% CI: 12%, 1405%) among those having any carpet in the home versus those with no carpet in the home. Participants with a doormat in the entryway of their home had significantly lower (p<0.05) concentrations of cypermethrin (−87%), Σpermethrin (−99%), fipronil sulfide (−69%) and DDE (−75%) than those without a doormat. Cypermethrin concentrations were more than nine times (936%) higher if participants reported having an exterminator at the house during the previous six months and concentrations of Σpermethrin were an estimated 23 times higher (2378%) if home-use pesticide products were present in the home during the sampling period, compared to no exterminator and no home-use pesticide products, respectively. Paradoxically, any self-reported pesticide use in the home during the past 6 months was associated with lower concentrations of Σpermethrin. Overall, the full Tobit models provided a moderate improvement over the intercept model (pseudo-R^2 = 0.25 - 0.44) with the greatest improvement for ∑permethrin concentrations.

4. Discussion

We detected multiple pesticides in personal monitoring wristbands worn by adolescent girls living in a California agricultural community, including pesticides currently used for agricultural and residential purposes, as well as legacy pesticides that have not been used for between 6 and 46 years. Doormats in home entryways and cleaning the home daily were associated with lower concentrations of certain chemicals, whereas carpeting in the home, having an exterminator treat the home in the past six months, and living within 100 m of active agricultural fields were associated with a higher odds of detecting some pesticides.

Several of the most frequently detected chemicals have been associated with potential adverse health effects. Fipronil, the parent insecticide of metabolites fipronil sulfide and fipronil sulfone, has exhibited oncogenicity and neurologic toxicity in animal studies, yet its implications for human health are unclear.[26] Cypermethrin, permethrin, and dacthal are all classified as possible carcinogens. Although human studies are few, epidemiologic evidence suggests pyrethroid pesticides may be associated with chronic health effects like immune system suppression and carcinogenesis.[27-28]

Silicone wristbands are a relatively new method of passive sampling that is nonintrusive and easy to use. A previous study in a heavily deforested region of Peru with developing agriculture measured pesticides in wristbands using the same methodology and found higher concentrations of chlorpyrifos, cypermethrin, and DDT in wristbands worn by participants living in agricultural compared to non-agricultural communities, and that community of residence and participant age explained about 40% of the variability in pesticide concentrations. In our study, we evaluated more predictors than the study in Peru and used Tobit regression to account for censoring of data below the LOD but were able to develop models for pesticide concentrations in bracelets that were significant improvements over intercept models (pseudo-R^2 = 25 - 44%). A higher percentage of wristbands from the study in Peru had detectable levels of chlorpyrifos (91% vs. 36%), DDT/DDE (97% vs. 56%) and cypermethrin (71% vs. 56%) than in our study, perhaps because participants in Peru wore the wristbands for 30 days compared to 7 days in our study (Table S4). In a previous study of wristbands worn for five days by farmworkers in Senegal, West Africa, more frequent detection was seen in West Africa than in our study for cypermethrin (94% vs. 56%) and chlorpyrifos (51% vs. 36%), while permethrin (27% vs. 55%) and DDE (37% vs. 56%) were detected more frequently in our study (Table S4)..[29]

Although few studies have been conducted on silicone wristbands, previous studies on pesticides in house dust confirm our findings that personal exposures are predicted by factors such as farmworkers in the home, proximity to agricultural crops, and pesticide storage in the home.[9,18,30] In house dust samples collected from 197 CHAMACOS residences, higher levels of chlorpyrifos, diazinon, dacthal and permethrin were found in house dust if farmworkers stored their shoes in the home; and higher levels of dacthal and iprodione were observed if there was agricultural use of these pesticides with ~2.7 km of the residence.[18] Passive indoor and outdoor air samples analyzed for chlorpyrifos and azinphos-methyl from 23 homes in Washington state found higher outdoor air concentrations of both pesticides among houses within 250 m of an apple orchard and higher indoor air concentrations of both pesticides in farmworker households compared to non-farmworker households. Indoor air concentrations of diazion and piperonyl butoxide measured in 102 homes in New York City were higher if participants reported using an exterminator, insecticide spray or bomb compared to those with no reported pest control applications.[31]

There are several strengths and limitations to our study. Although this is the largest study to date using wristbands to measure personal exposure to pesticides, the sample size in our study was still fairly small with fewer than 100 participants. We evaluated numerous combinations of predictors and pesticide concentrations, and given our sample size, none of our results would be significant if adjusted for multiple comparisons. Additionally, few pesticides were detected in more than 50% of wristbands, making it difficult to compare concentrations across participants for several pesticides, and limiting some statistical analyses to simply the presence or absence of the target analyte.

Pesticide concentrations in wristbands can reflect potential exposure through inhalation and dermal routes, but do not reflect exposure from ingestion. [11,22] Although pesticides measured in wristbands demonstrate potential exposure, it should not be considered equivalent to internal dose due to differences in uptake between silicon wristbands and humans.

Importantly, the analytical methods and silicone wristband matrix influence which pesticides are detected. The measured concentrations of different pesticides are not only attributable to the presence of these pesticides in the environment, but also are likely to depend upon environmental conditions like temperature and relative humidity and physical properties of the pesticides such as vapor pressure and the partition coefficient between silicone and air.[22-23]

However, uptake factors should be consistent for each individual pesticide across the sampling devices allowing comparison of concentrations of the same pesticide between wristbands.

We collected extensive information on home pesticide use and housing characteristics. Although many of the housing characteristics were collected by objective observation, other factors (including frequency of house cleaning, recent aerial spraying, and how often windows were open at night) relied on subjective self-report. Questionnaires were asked of the adolescent participants who may have been less familiar with some items, such as how often pesticides were used in their homes or how often their homes were cleaned, than their parents. None-the-less, despite this potential for misclassification, we were able to identify predictors of pesticide concentrations in wristbands.

Our study had very good compliance, demonstrating that silicone wristbands can be used for personal sampling of adolescents. While we only included girls in the current study, it is possible that some results might differ by participant gender, particularly if boys were more involved in farm work.This study took a unique approach to environmental health research by involving YRAs in its design, recruitment, and data collection. As the YRAs worked closely with study leaders, it is unlikely that any error in data collection was introduced by using this community-based model. Overall, we found that questionnaires, field

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identification, and sample collection were improved by the involvement of community informants.

5. Conclusions

In this study, we detected multiple pesticides in silicone wristbands worn for one week by Latina teenage girls living in the agricultural Salinas Valley in California. Although we detected several agricultural pesticides, many of the pesticides we detected were used only for residential pest control or were legacy pesticides that had not been used for many years, suggesting that agriculture is not the only source of pesticide exposure in this community. Nearby agricultural pesticide use, home pesticide treatments, and individual behaviors like cleaning the home daily and having a doormat in the entryway were associated with levels of several pesticides in wristbands, and some of these results were consistent with past studies using wristband, air, or dust samples.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Highlights

- Multiple pesticides were detected in silicone personal monitoring wristbands.
- Dacthal and permethrin concentrations were 3x higher among girls living near fields.
- Pesticide levels were higher if pesticides had been used or stored in the home.
- Levels were lower among girls with homes that had door mats or were cleaned daily.

Table 1.

Characteristics, instrument limits of detection and percentage detected for pesticides frequently detected in silicone wristbands in the COSECHA study (n=97).

Pesticide	Type	Uses[a]	Agricultural use in Monterey 2016 (kg)[b]	Instrument LOD[c] (Pg/^L)	Detected (%)
Current Agricultural Use					
Dacthal (chlorthal dimethyl)	Herbicide	Broccoli, onions, cauliflower	29,910	1.34	52.6
Chlorpyrifos	Insecticide	Grapes, brussel sprouts, strawberries	1,460	1.82	36.1
Dimethoate	Insecticide	Celery, broccoli, lettuce	3,500	1.78	13.5
Current Agricultural and Residential Use					
Cypermethrin	Insecticide	Lettuce, broccoli, artichokes: ants, roaches	1,520	5.08	55.7
trans-Permethrin[d]	Insecticide	Lettuce, spinach, celery: ants, roaches, lice	14,860	1.27	51.5
cis-Permethrin[d]	Insecticide	Lettuce, spinach, celery: ants, roaches, lice	14,860	0.73	48.5
Esfenvalerate	Insecticide	Broccoli, artichokes; broad spectrum	790	6.11	41.2
Ethoprop (Prophos)	Insecticide	Cabbage; lawns	14	22.2	34.0
Oxadiazon	Herbicide	Flowers; lawns	28	2.95	20.8
Piperonyl butoxide	Synergist	Strawberries, flowers; ants, roaches	1,510	NA	19.2[e]
Phosmet (Imidan)	Insecticide	Potted plants; fleas/ticks	37	1.55	10.4
Current Residential Use					
Fipronil sulfide	Breakdown Product	Breakdown product of Fipronil	NA	3.09	86.6
Fipronil sulfone	Breakdown Product	Breakdown product of Fipronil	NA	2.93	45.4
Fipronil	Insecticide	Broad spectrum, fleas/ticks	0	4.36	10.4
DEET[f]	Insect Repellant	Mosquitoes	0	NA	21.2[e]
Historical Use					
DDE	Breakdown Product	Banned 1972, breakdown product of DDT	0	1.46	55.7
Propachlor	Herbicide	Discontinued 1998	0	5.72	53.6
Ethion	Insecticide	Citrus; lawns, last used in MC[g] 1992	0	5.23	39.2
Chloroneb	Fungicide	Soybeans; lawns, last used in MC[g] 1999	0	9.86	34.0
Dicofol	Miticide	Cotton, citrus, last used in MC[g] 2012	0	20.9	33.0

Pesticide	Type	Uses[a]	Agricultural use in Monterey 2016 (kg)[b]	Instrument LOD[c] (Pg/^L)	Detected (%)
Dieldrin	Insecticide	Banned 1987	0	0.55	22.9
Endrin aldehyde	Breakdown Product	Banned 1991	0	0.44	21.9
trans-Nonachlor	Component	Banned 1988, component of chlordane	0	1.08	14.6
α-Chlordane	Insecticide	Banned 1988	0	1.02	12.5
Mirex	Insecticide	Banned 1978	0	1.48	12.5

[a] Most common crops for Monterey County.

[b] California Pesticide Use Report Data 2016.

[c] Instrument limit of detection on the quantitative pesticide method, determined over three analytical days (n=7). LODs not available for DRS screening method.

[d] Total permethrin.

[e] Present based on DRS screening method.

[f] DEET = N-N-diethyl-m-toluamide.

[g] MC = Monterey County.

Table 2.

Detection frequencies and distributions of most commonly detected pesticides (>33% detection) in wristbands (n=97).

		Concentration (ng/g/day)				
Pesticide	**Detect (%)**	**Mean**	**SD**	**p50**	**p75**	**p95**
Fipronil sulfide[a]	86.6	34.5	49.0	12.9	36.6	145
DDE	55.7	1.1	1.1	0.7	1.4	3.2
Cypermethrin	55.7	13.9	38.8	2.3	8.4	116
ΣPermethrin	54.6	154	1 010	7.6	29.5	494
Propachlor	53.6	5.4	7.2	2.3	8.3	20.3
Dacthal	52.6	1.7	5.9	0.3	1.7	5.3
Fipronil sulfone[a]	45.4	0.7	0.9	<LOD	0.7	2.7
Esfenvalerate	41.2	8.1	41.9	<LOD	3.2	32.4
Ethion	39.2	1.2	1.6	<LOD	1.6	4.4
Chlorpyrifos	36.1	0.5	1.0	<LOD	0.6	2.1
Chloroneb	34.0	14.1	42.0	<LOD	13.2	51.6
Ethoprop	34.0	5.1	9.7	<LOD	5.1	19.0
Dicofol	33.0	1.2	1.7	<LOD	0.9	3.4

[a]Breakdown products of fipronil.

Table 3:

Unadjusted odds ratios (OR) and 95% confidence intervals (CI) for presence of pesticides in wristbands (n=97).

	Agricultural Use				Agricultural and Residential Use						Historical Use	
	Dacthal		Chlorpyrifos		∑Permethrin		Esfenvalerate		Piperonyl butoxide[a]		DDE	
Characteristic	**OR**	**(95% CI)**	**OR**	**(95% CI)**	**OR**	**(95% CI)**	**OR**	**(95% CI)**	**OR**	**(95% CI)**	**OR**	**(95% CI)**
Crops within 100m of home	3.1	(1.0, 9.5)*	1.0	(0.4, 3.0)	2.1	(0.7, 6.0)	0.8	(0.3, 2.2)	1.2	(0.4, 4.2)	2.7	(0.9, 8.1)†
Agricultural workers at home	1.6	(0.7, 3.7)	1.1	(0.4, 2.8)	0.8	(0.3, 1.9)	0.5	(0.2, 1.3)	0.3	(0.1, 1.0)*	0.6	(0.2, 1.4)
Aerial spraying seen nearby	2.0	(0.9, 5.5)	0.9	(0.4, 2.2)	1.2	(0.5, 2.8)	0.9	(0.4, 2.1)	1.5	(0.6, 4.3)		NA
Lives in Salinas	0.2	(0.1, 0.4)*	0.9	(0.4, 2.1)	1.2	(0.5, 2.6)	1.1	(0.5, 2.5)	0.6	(0.2, 1.8)	0.9	(0.4, 2.0)
Window open at night[b]	2.0	(0.9, 4.5)†	0.7	(0.3, 1.7)	1.2	(0.5, 2.6)	2.7	(1.2, 6.5)*	0.9	(0.3, 2.4)		NA
Carpet in home	3.7	(1.4, 10.0)*	1.2	(0.4, 3.1)	0.8	(0.3, 2.1)	1.6	(0.6, 4.1)	1.8	(0.5, 6.9)	1.7	(0.7, 4.3)
Doormat present[c]	1.7	(0.5, 6.6)	0.5	(0.1, 1.9)	0.1	(0.1, 0.9)*	1.7	(0.4, 6.9)	0.2	(0.1, 0.7)*	0.1	(0.1, 1.0)†
Clean daily	0.8	(0.4, 1.9)	1.1	(0.4, 2.5)	0.4	(0.2, 1.0)†	0.8	(0.4, 1.9)	0.3	(0.1, 0.8)*	1.4	(0.6, 3.3)
Insecticide use, last 6 mo		NA		NA	0.3	(0.1, 0.8)*	0.6	(0.3, 1.5)	0.6	(0.2, 1.8)		NA
Exterminator, last 6 mo		NA		NA	6.5	(0.8, 55.4)†	4.9	(0.9, 25.4)†	9.7	(2.1, 45.8)*		NA
Pesticides in home		NA		NA	2.9	(1.3, 6.6)*	0.6	(0.3, 1.3)	0.6	(0.2, 1.6)		NA
Pets in home		NA		NA	1.4	(0.6, 3.2)	1.4	(0.6, 3.2)	0.9	(0.3, 2.5)		NA

[a]Odds Ratios for presence of pesticide in wristbands;

[b]at least once that week;

[c]n = 96;

†p< 0.1;

*p<0.05.

Table 4.

Bivariate analysis of determinants of continuous pesticide levels (ng/g/day) in wristbands (n=97) using the Wilcoxon rank sum test.

			Geometric Mean (ng/g/d)					
Characteristic	Detect	%	Dacthal	Cypermethrin	∑Permethrin	Fipronil sulfide	DDE	Propachlor
Crops within 100 m of home	Yes	19.6%	1.0	1.4	8.6	NA	0.8	1.7
	No	80.4%	0.3*	2.3	2.3*		0.4*	1.9
Agricultural worker in home	Yes	70.1%	0.5	2.3	2.4	NA	0.4	2.1
	No	29.9%	0.3	1.7	4.4		0.5	1.5
Aerial spraying seen nearby	Yes	41.2%	0.5	1.4	3.0	NA	NA	NA
	No	58.8%	0.3	2.8*	2.9			
Lives in Salinas	Yes	58.8%	0.2	2.3	3.2	11.3	0.4	2.4
	No	41.2%	1.1**	1.9	2.6	8.1	0.4	1.6
Window open at night[a]	Yes	58.8%	0.5	2.4	3.2	NA	NA	NA
	No	41.2%	0.3	1.7	2.6			
Carpet in home	Yes	75.3%	0.5	2.2	3.0	10.5	0.5	2.1
	No	24.7%	0.2†	1.7	2.7*	8.0	0.3	1.3†
Doormat present[b]	Yes	89.6%	0.4	1.9	2.3	9.5	0.4	1.8
	No	10.4%	0.3	4.7	21.7*	11.6	0.7	2.5
Clean daily	Yes	65.0%	0.4	2.1	1.9	11.7	0.5	1.8
	No	35.1%	0.4	2.0	6.8*	7.2	0.3	2.2
Insecticide use, last 6 mo	Yes	58.8%	NA	6.8	1.9	7.3	NA	NA
	No	41.2%		1.9†	5.3†	15.2†		
Exterminator, last 6 mo	Yes	8.3%	NA	6.8	22.7	13.0	NA	NA
	No	91.8%		1.9	2.4*	9.6		
Pesticides in home	Yes	51.0%	NA	2.3	6.9	10.0	NA	NA
	No	49.0%		1.9†	1.2*	9.7		
Pets in home	Yes	52.6%	NA	2.3	3.3	9.6	NA	NA
	No	47.4%		1.8	2.6	10.1		

[a] at least once that week;

[b] n = 96;

† $p < 0.1$;

* $p<0.05$;

** $p<0.001$.

Table 5.

Percent change[a] in pesticide levels (ng/g/d) in wristbands from multivariable models (n=96).

Characteristic	Dacthal		Cypermethrin		ΣPermethrin		Fipronil sulfide		DDE		Propachlor	
	%	95% CI	%	95% CI	%	95% CI	%	95% CI	%	95% CI	%	95% CI
Crops within 100 m of home	73	(−56, 575)	−70	(−95, 85)	1194	(−5, 17436) †	NA		191	(−8, 817) †	−32	(−83, 178)
Agricultural worker in home	84	(−38, 444)	75	(−60, 663)	80	(−78, 1367)	NA		−38	(−77, 68)	67	(−57, 543)
Aerial spraying seen nearby	−18	(−73, 146)	−68	(−92, 33)	−59	(−96, 276)	NA		NA		NA	
Lives in Salinas	−93	(−97, −84) **	−9	(−77, 250)	425	(−35, 4169)	80	(−29, 359)	34	(−49, 250)	−62	(−88, 21)
Carpet in home	311	(12, 1405) *	113	(−52, 840)	−17	(−89, 510)	30	(−53, 261)	127	(−24, 577)	63	(−54, 474)
Doormat present	180	(−48, 1398)	−87	(−98, −24) *	−99	(−100, −80) *	−69	(−88, −17) *	−75	(−91, −32) *	−36	(−87, 222)
Clean daily	−30	(−78, 120)	−5	(−78, 299)	−49	(−93, 292)	52	(−40, 286)	153	(−9, 605) †	−25	(−77, 142)
Insecticide use, last 6mo	NA		−30	(−82, 166)	−94	(−99, −67) *	−71	(−89, −28) *	NA		NA	
Exterminator, last 6mo	NA		936	(79, 5912) *	330	(−81, 9717)	−5	(−73, 235)	NA		NA	
Pesticides in home	NA		47	(−60, 440)	2378	(275, 16271) **	44	(−40, 249)	NA		NA	
Overall model pseudo-R^2		0.42		0.34		0.44		0.33		0.39		0.25

[a] Percent change = [exp(β)−1]*100;

† $p < 0.1$;

* $p < 0.05$;

** $p < 0.001$.