Author's response to reviews

Title: Pesticides in dust from urban and farmworker households in California: an observational study.

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Author’s response to reviews: see over
Dear Environmental Health Editorial Board:

We appreciate the reviewers’ review of our manuscript now entitled, “Pesticides in dust from urban and farmworker households in California: an observational study.” Below are our point-by-point responses to the reviewers’ comments. A new version of the manuscript has been submitted with the revisions in “tracked changes”.

Editor’s Suggested Revisions:

Per revisions suggested by the editor, the following changes have been made:

(1) **Editorial comment:** Please remove the italics throughout the manuscript text, except where instructed i.e. in the references.

**Response:** All italicized words in the document have been revised to be in regular font.

(2) **Editorial Comment:** On the title page please state the study design after the title e.g. X is a risk factor for Y: a case control study and a line break is needed between the first and second institutional addresses as the other already do.

**Response:** The title has been revised to reflect the type of study design as follows:

Old title: Pesticides in dust from urban and farmworker households in California  

Revised title: Pesticides in dust from urban and farmworker households in California: an observational study.

Additionally, a line break between the first and second institutional addresses has been inserted.

(3) **Editorial Comment:** The abstract, like the rest of the manuscript, should be formatted with double line spacing.

**Response:** The abstract has now been formatted to be double-spaced.

(4) **Editorial Comment:** In the references section, the issue numbers should be removed.

**Response:** Issue numbers have been removed.

(5) **Editorial Comment:** All the horizontal lines in the tables should be visible.

**Response:** Horizontal lines have been added to all tables including supplemental tables.

(6) **Editorial Comment:** All tables in landscape format must be reformatted onto a portrait page or submitted as additional files. (Any additional files will be linked into the final published article in the form supplied by the author, but will not be displayed within the paper. They will be made available in exactly the same form as originally provided.)

**Response:** All tables in landscape format have been reformatted onto a portrait page.
Reviewer: Dr. Linda McCauley

(1) Reviewer Comment: Suggest that more citations be included in background section on researchers that have measured dust in residential areas. Including only 3 citations under-represents the substantial work that has been done in this area.
Response: The authors have added five citations: Butte & Heinzow 2002, Colt et al. 2004, Rudel et al. 2003, McCauley et al. 2001, and Simcox et al. 1995. Additionally, one of the original citations (Roberts et al. 2009) and Butte & Heinzow 2002 are review articles that include other relevant citations.

(2) Reviewer Comment: Background last paragraph…change to “Finally, we estimated”
Response: Edit suggested by reviewer has been made.

(3) Reviewer Comment: Page 5, under dust sample collection. Concern about including two homes where furniture was sampled instead of floors. Need to know if excluding those samples changes the findings that are reported.
Response: The authors have included the following paragraph in the results section under “Dust levels: Trends and location differences”

“Dust concentrations from furniture samples in two farmworker homes were comparable to those collected from carpets in other farmworker homes for frequently detected OPs (diazinon and chlorpyrifos), piperonyl butoxide, and chlorthal-dimethyl, while for frequently detected pyrethroids, concentrations were generally at the upper end of the distribution. We observed the same general pattern when using loadings. Maximum permethrin concentrations in farmworker homes were observed in furniture samples; however, the highest permethrin concentrations were observed in carpet samples from urban homes. The highest loading observed for cypermethrin was collected from a furniture sample; however, higher loadings were observed in carpet samples from urban homes. No location differences in pesticide concentrations or loadings were observed when we excluded furniture samples from our analysis.”

The authors have also added a footnote to Tables 2 and S2 to indicate that samples from two farmworker homes were collected from furniture due to the absence of a carpet/area rug in each home.

(4) Reviewer Comment: Page 5, on second day sampling….need to know how the researchers knew they were not vacuuming in the same areas previously sampled. Needs to be close, but not the same sampled area.
Response: As stated in the methods section, dust samples were obtained from the same general location. This was done based on recall from study personnel collecting the samples. The intent of our study design was not to evaluate within-home spatial variability, but rather stability of levels in the same general area over time. No changes have been made to the manuscript.

(5) Reviewer Comment: A large proportion of the homes had insufficient sample mass for analysis. This needs to be addressed as a weakness. Could present a bias into the findings.
Response: The authors have included the following statement in the last paragraph of the discussion which details study limitations:

“Additionally, although homes with insufficient sample mass were demographically similar to those with adequate sample mass, exclusion of these homes may have introduced some bias and prevented us from detecting a difference in pesticide concentrations and/or loadings between locations.”
(6) **Reviewer Comment:** On page 8, would be helpful to describe the demographic and household variables that were measured in 2002 and the current study.

**Response:** The authors have revised the text to indicate the demographic/household characteristics as follows:

There were no demographic or household differences between our previous studies and the present study; i.e., all households had at least one farmworker residing in the home and study participants generally represented the farmworker population in Salinas Valley which is primarily from Mexico or of Mexican descent, low –literacy, Spanish-speaking, low-income, frequently reported pesticide applications indoors, and had most farmworker residents wear their work clothing indoors. Homes were also located >200 feet from the nearest agricultural field.

(7) **Reviewer Comment:** On page 10, what were the pyrethroid insecticide levels in homes reporting recent application? Were they outliers?

**Response:** The following statement has been added under the “Dust levels: trends and location differences” in the results section.

“Cypermethrin was applied in one farmworker home, while imiprothrin was applied in two urban homes between the two sampling dates. For the farmworker home, cypermethrin dust concentrations were at the upper end of the distribution among other farmworker homes (between the 75th and 95th percentile concentrations reported). Imiprothrin was only detected in one of the urban homes which reported usage during the study; no other urban home had detectable imiprothrin levels indoors even though some of these households reported applying imiprothrin indoors prior to the study.”

(8) **Reviewer Comment:** Interesting that the urban and rural homes did not differ significant on their loadings. This is very different from findings from other investigators and needs to be pointed out. Was this primarily a limitation of sample size?

**Response:** We did not observe statistically significant differences between locations for concentrations or loadings as indicated in the manuscript. It is possible that this may have resulted from the small sample size.

We have revised the limitations paragraph to indicate this as follows:

“This study has several limitations. Location differences in pesticide dust levels have been reported previously when using loadings rather than concentrations [21]; however, our small sample size limits statistical power and may have prevented us from observing statistically significant differences between locations for concentrations and/or loadings. Additionally, although homes with insufficient sample mass were demographically similar to those with adequate sample mass, exclusion of these homes may have introduced some bias and prevented us from detecting a difference in pesticide concentrations and/or loadings between locations.”

(9) **Reviewer Comment:** Need to discuss the higher levels of malathion in urban homes. Isn’t this an agricultural chemical or used for outdoor pest control?

**Response:** The authors have included the following statements in the results section:

“Malathion was not frequently detected in homes from either location; however, higher levels were observed in urban homes. This pesticide is used in agriculture and is also registered for use in home gardens, as a building perimeter treatment, as a wide-area spray for mosquitoes, and by prescription for head-lice control [36]. However, no parents reported treating their children for lice or using it themselves in their gardens. The main county uses for this OP pesticide in 2006 in the urban region were landscape maintenance and..."
These applications were reported more than 25 km away from the nearest study home, thus it is not readily apparent why higher levels were observed in urban homes although it should be noted that we only sampled a small number of homes.”

Reviewer: Dr. Nicolle Tulve

(1) **Reviewer Comment:** The manuscript is missing some of the current relevant literature related to dust concentrations. Incorporation of the most current literature is recommended.

**Response:** References have been added throughout the manuscript including: Curl et al. 2002, Colt et al. 2004, Fenske et al. 2002, Rothlein et al. 2006, Rudel et al. 2003, McCauley et al. 2001, and Simcox et al. 1995

(2) **Reviewer Comment:** In addition to focusing on a subset of the CHAMACOS cohort for comparison purposes (discussion section primarily), the manuscript would benefit from a more wholesale comparison of the dust concentrations that have been reported for pesticides related to other exposure assessments, especially in regards to other rural/agricultural cohorts. This would probably help to put the non-dietary ingestion exposure estimates in perspective. Consider expanding the discussion section and including a comparison table.

**Response:** The authors have included the following paragraph/revisions in the discussion section and have made a new table (Table 6) describing dust concentrations in select farmworker studies:

“Compared to other studies in farmworker populations (Table 6), we observed lower median concentrations for chlorpyrifos [10, 13, 22, 38, 39] and diazinon. These farmworker studies also reported a wider range of concentrations for these two OP pesticides and generally collected dust samples prior to the residential phase-out. One study by Curl et al. [22] reported a wider range of diazinon concentrations, but comparable median concentrations (10 ng/g). Although malathion was not frequently detected in our farmworker homes, a wider range of concentrations was reported in previous farmworker studies (Table 6) [22, 39]. To our knowledge, only one other study has reported OP pesticide concentrations in low-income urban homes [40]. This study reported higher median concentrations for chlorpyrifos and diazinon in low-income urban housing units in Boston, MA. Homes in this study were sampled just after or during the residential phase-out of chlorpyrifos and diazinon, respectively (between July 2002 and August 2003).”

“Pyrethroids were detected in house dust in several study homes. Similar to low-income urban housing units in Boston, MA [30], pyrethroids and PBO were detected in higher concentrations and used more frequently in our study homes compared to other pesticides. This finding is consistent with the fact that pyrethroid insecticide formulations for residential applications have largely replaced OP pesticide residential formulations [31, 32]. Although over 19,000 kgs of permethrin were applied in Monterey County in 2006 for agricultural purposes [33], we did not observe significant differences in permethrin concentrations between locations. Allethrin and cypermethrin were also widely detected in most homes. Our findings suggest that home use likely contributed to the presence of pyrethroid pesticides in house dust since pyrethroids were commonly used indoors and negligible to no agricultural applications took place at the county level (except for permethrin). It is also possible that structural pest control applications influenced indoor detection of certain pyrethroids in some homes. For example, it is estimated that ~80% of the non-agricultural cypermethrin use reported in Alameda County in 2006 was for structural pest control [34]. The presence of pyrethroids in house dust is also consistent with their physical and chemical properties, including high octanol:water partition coefficient values (log $K_{ow} > 4.0$) and
low vapor pressures (SI, Table S1). To our knowledge, only two studies [10,13] have measured pyrethroid dust concentrations in farmworker homes. Similar to the present study, permethrins were the most frequently detected pyrethroids indoors. Median cis- and trans-permethrin concentrations in our farmworker homes were higher than those observed in previous farmworker homes [10].

(3) **Reviewer Comment:** There are a limited number of literature articles that suggest that high socio-economic status relates to high pesticide residues in the home. This would mean that these homes may have higher dust concentrations. Consider how this issue will be acknowledged in the manuscript.

**Response:** The authors are not aware of such literature. No changes have been made at this time.

(4) **Reviewer Comment:** Page 3, third paragraph, there is a discussion of semi- and non-volatile pesticides. Chlorpyrifos, as a recognized semi-volatile pesticide, behaves much differently than the dust-bound pesticides (i.e., pyrethroids), the non-volatile pesticides. Consider separating the discussion of semi-volatile and non-volatile pesticides to clearly articulate how their physico-chemical properties affect their behavior in the indoor environment and where they may be found (e.g., air for chlorpyrifos, dust-bound for pyrethroids).

**Response:** The following revisions have been made:

“Several studies indicate that pesticide residues persist indoors due to the lack of sunlight, rain, temperature extremes, microbial action, and other factors that facilitate degradation [12]. Semi- and non-volatile pesticides (e.g., OPs and pyrethroids) have chemical properties that increase binding affinity for particles and the tendency to adsorb onto household surfaces such as carpet or dust, also prolonging their persistence indoors [16]. For example, pyrethroid pesticides have low vapor pressures, and high octanol/water (Kow) and water/organic carbon (Koc) partition coefficients which facilitate partitioning into lipids and organic matter and binding to particulate matter in dust. Human exposure results from particle-bound movement and transfer [21]. In fact, several studies suggest that house dust is an important pathway of pesticide exposure for children [11, 15, 17, 22]. Young children are particularly vulnerable to inadvertent ingestion of pesticide-contaminated dust due to their frequent hand-to-mouth activity and contact with indoor surfaces [12].”

(5) **Reviewer Comment:** Page 3, third paragraph, a reference is needed for the sentence “Several studies suggest that house dust is an important pathway of pesticide exposure for children.”

**Response:** The authors have added the following references to this statement: Butte and Heinzow 2002, Curl et al. 2002, Roberts et al. 2009, and Simcox et al.1995

(6) **Reviewer Comment:** The inclusion of dust samples analyzed several days apart, the explanation for why this was done and why it advances the scientific literature, is weak. The scientific community seems to accept that dust is both a source and sink for contaminants, and that the concentration is fairly stable, especially when sampled only a few days apart. I’m not sure I understand the added benefit or the scientific advance in including this portion of the manuscript. Consider how to reframe this part to improve the overall need to include it.

**Response:** The authors feel that it is important to comment on whether the samples were correlated within homes. The following revision has been made in the discussion section:

“We detected several pesticides in most homes, including OP pesticides previously phased-out for residential uses, pyrethroids, and the pesticide synergist piperonyl butoxide (PBO). Biological exposure metrics for these pesticides are relatively transient and highly variable, typically reflecting recent exposures [29]. However, consistent with other studies [15, 35], we found that dust serves as a stable matrix and indicator of potential indoor
exposure for some pesticides in house dust.” The high correlations observed in dust concentrations from samples collected 5-8 days apart suggests that, for some pesticides, measurements in house dust may be relatively stable indicators of potential indoor exposure over this time frame.

(7) Reviewer Comment: Consider whether you need to include the discussion of the pesticide related survey questions that were asked since they are not included in any of the analyses presented in the manuscript. Or, if there is strong desire to include this discussion, expand on it in the results and discussion sections of the manuscript. A data gap in the scientific literature is understanding how survey questions and responses can be used to predict pesticide concentrations in the home a useful manuscript to discuss these implications and needs.

Response: The authors have added the following statements in the results and discussion sections, respectively:

Results:
“Most participants reported using pesticides indoors in the three months preceding the study (67% and 85% of farmworker and urban homes, respectively) and the most common location of use was the kitchen.”

Although not statistically significant, we generally observed higher dust concentrations in homes that reported recent pesticide use (i.e., within the last three months preceding the study) when pesticide containers were available to confirm the active ingredients.

Discussion:
“Although we surveyed participants on their usage of pesticides indoors, we were not always able to corroborate whether formulation ingredients were present at high concentrations as the pesticide containers were not always available to confirm the active ingredients.”

The authors feel that it is important to state the results from the survey questions, however due to the absence of some data (as stated in the added discussion statement) we were not always able to predict pesticide concentrations indoors and feel that the information collected is not sufficient to comment on understanding “how survey questions and responses can be used to predict pesticide concentrations in the home.”

(8) Reviewer Comment: Two homes had samples collected from the furniture because there was no rug in the home from which to sample. Consider whether these samples are comparable to the floor samples. Consider how this should be described and documented in the manuscript.
Response: Another reviewer raised the same concern. Please see response #3 under previous responses to the Dr. Linda McCauley.

(9) Reviewer Comment: Portions of the description of the multi-residue analysis method appear very similar to previously published multi-residue analysis methods for soil and surface wipes. If the method has been adapted/modified from a previously published method, consider how proper acknowledgement and citation will be provided in the manuscript.
Response: We have revised the lab section to include two references (Bradman et al. 2006 and Harnly et al. 2009) as follows:
“To measure analytes, we modified a previously used laboratory method [10, 13]. Briefly, 0.5 g dust aliquots were fortified with 250 ng of two surrogate recovery standards (SRSs)--fenchlorphos and $^{13}$C$_{12}$-trans-permethrin--and extracted using ultrasonication in 1:1 hexane:acetone…..”
Reviewer Comment: The conclusion stating “…there is a need to educate families on the potential health impacts of pesticide use and effective integrated pest management strategies to control pests” (presented in both the end of the abstract and on page 15) does not reflect the results and discussion in the manuscript. Consider some discussion of how education strategies within the context of understanding dust concentrations in homes would improve pest control strategies, reduce exposures of family members, and better enable understanding of dust concentrations.

Response: The authors feel that the high prevalence of pesticide usage reported by households in both locations supports this conclusion. Educating families on the potential health effects of pesticides may serve as an incentive to eliminate the use of pesticides indoors and try safer methods of pest control. In fact, the study cited (Williams et al. 2006) found that integrated pesticide management (IPM) is an effective strategy for reducing pest infestation levels indoors and a person’s internal insecticide dose.

We have also made the following revisions in the abstract and conclusion section:

Abstract (results section):
“More than half of the households reported applying pesticides indoors.”

Conclusion:
“The frequent pesticide use reported among participating households in this and previous studies of low-income homes [13, 14, 30] and high detection of several pesticides in house dust suggests there is a need to educate families on the potential health impacts of pesticide use and effective integrated pest management strategies to control pests and reduce exposures to household occupants [31].”

Minor Essential Revisions:

(1) Reviewer Comment: Page 2, chlorthal-dimethyl is misspelled in the second paragraph.
Response: The correct spelling has been incorporated.

(2) Reviewer Comment: Page 9, the proper name of FQPA is the “Food Quality Protection Act”.
Response: The correct spelling has been incorporated.

(3) Reviewer Comment: Consider significant figures when presenting the data in Table 2; Consider significant figures when presenting the data in Table S2.
Response: Values on Tables 2 and S2 have been corrected to reflect 3 significant figures.

Discretionary revisions:

(1) Reviewer Comment: Consider including how assent of the children was obtained.
Response: Assent of children was not required by the UC Berkeley Center for Protection of Human Subjects so long as we obtained written informed consent from parents given the age of the children participating in the study.

(2) Reviewer Comment: Consider not including diazinon-oxon in Table S1 since there are no p-chem data inputs that can be provided.
Response: Diazinon-oxon and its accompanying footnote have been removed from Table S1.

Please note that the following minor revisions were also made upon revision of the new manuscript draft. These edits do not change our results or conclusions.
Abstract (results section):
“Analytes frequently detected in both locations included chlorpyrifos, diazinon, permethrin, allethrin, cypermethrin, and piperonyl butoxide; no differences in concentrations or loadings were observed between locations for these analytes.”

Background section:
“Due to their potential health effects in children, formulations of the OP insecticides chlorpyrifos and diazinon were voluntarily phased out for residential uses in 2001-2004 between 2001 and 2004.”

“We also examined whether house dust concentrations of chlorpyrifos and diazinon declined in Salinas, CA after the U.S. Environmental Protection Agency’s (EPA) voluntary residential phase-out of these compounds.”

Conclusion:
“A particular at-risk population are households with poorer housing quality, where there may be greater needs for pest control.“

“Particular at-risk populations are those living in households with poorer housing quality, where there may be greater needs for pest control. “

We thank the reviewers and editors of Environmental Health for their careful review of our manuscript and for considering it for publication. Future correspondence should be addressed to the main authors, Drs. Asa Bradman (abradman@berkeley.edu) and Lesliam Quiros Alcala (lquiros@berkeley.edu).

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