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Hazard-Ranking of Agricultural Pesticides for Chronic Health Effects in Yuma County, Arizona

Anastasia J. Sugeng1,* , **Paloma I. Beamer**1, **Eric A. Lutz**1, and **Cecilia B. Rosales**¹ ¹Community, Environment and Policy, Mel and Enid Zuckerman College of Public Health, University of Arizona, 1656 East Mabel Street Tucson, AZ 85724

Abstract

With thousands of pesticides registered by the United States Environmental Protection Agency, it not feasible to sample for all pesticides applied in agricultural communities. Hazard-ranking pesticides based on use, toxicity, and exposure potential can help prioritize community-specific pesticide hazards. This study applied hazard-ranking schemes for cancer, endocrine disruption, and reproductive/developmental toxicity in Yuma County, Arizona. An existing cancer hazardranking scheme was modified, and novel schemes for endocrine disruption and reproductive/ developmental toxicity were developed to rank pesticide hazards. The hazard-ranking schemes accounted for pesticide use, toxicity, and exposure potential based on chemical properties of each pesticide. Pesticides were ranked as hazards with respect to each health effect, as well as overall chronic health effects. The highest hazard-ranked pesticides for overall chronic health effects were maneb, metam sodium, trifluralin, pronamide, and bifenthrin. The relative pesticide rankings were unique for each health effect. The highest hazard-ranked pesticides differed from those most heavily applied, as well as from those previously detected in Yuma homes over a decade ago. The most hazardous pesticides for cancer in Yuma County, Arizona were also different from a previous hazard-ranking applied in California. Hazard-ranking schemes that take into account pesticide use, toxicity, and exposure potential can help prioritize pesticides of greatest health risk in agricultural communities. This study is the first to provide pesticide hazard-rankings for endocrine disruption and reproductive/developmental toxicity based on use, toxicity, and exposure potential. These hazard-ranking schemes can be applied to other agricultural communities for prioritizing community-specific pesticide hazards to target decreasing health risk.

Keywords

pesticide prioritization; hazard-ranking; community health; chronic health effects

1. INTRODUCTION

Pesticide exposure is a major environmental health issue in agricultural communities (Arcury et al., 2006; Coronado et al., 2011; Liebman et al., 2007; Simcox et al., 1995). Cancer, endocrine disruption, and reproductive/developmental toxicity are among the health effects associated with chronic exposure to pesticides (Baldi et al., 2001; Garry et al., 2002;

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^{*}Corresponding author: Anastasia Sugeng, MS, 1656 East Mabel Street Tucson, Arizona 86724, Phone: (520) 901-0139, Fax: (520) 626-8009, asugeng@email.arizona.edu.

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Oliva et al., 2001; Zheng et al., 2001). Children are particularly vulnerable to these health effects due to their less-developed metabolism and the on-going maturation of their organ systems (Anderson et al., 2000; Faustman et al., 2000; Ribas-Fito et al., 2006).

The ever-increasing and ever-changing use of pesticides has made it unfeasible to rely on environmental and biological sampling for all pesticides to assess health risk in communities (Blasco and Picó, 2009). Hazard-ranking schemes have long been used to rank and prioritize chemicals of greatest health concern (Bro-Rasmussen and Christiansen, 1984; Schmidt-Bleek et al., 1982). Previous approaches to hazard-ranking environmental contaminants have often been limited by incomplete databases, requiring researchers to base hazard-rankings on "likely present contaminants" rather than on quantified levels of identified contaminants (Baun et al., 2006; Mitchell et al., 2002; Siljeholm, 1997). Another common approach has used toxicity to assess known chemical releases; however, consideration of exposure potential has often been lacking (Dix et al., 2007; Neumann et al., 1998). Finally, other hazard-rankings have focused on acute toxicity, neglecting chronic exposure and its associated health effects (Swanson et al., 1997; USEPA, 1992). Identifying and prioritizing pesticides with hazard-ranking schemes that consider pesticide use, toxicity for chronic health effects, and exposure potential is necessary for a more accurate assessment of health risk in agricultural communities.

Gunier (2001) has been a leader in the effort to improve pesticide hazard assessments by developing a pesticide hazard-ranking scheme applied to California for the cancer endpoint. Pesticide use was quantified in California between 1991 and 1994 from the Pesticide Use Report (PUR) system provided by the California Department of Pesticide Regulation. The method developed by Gunier (2001) incorporated exposure potential and cancer toxicity to calculate hazard factors, which were in turn used to calculate hazard-adjusted use for each pesticide. The top hazard-adjusted use pesticides in California from 1991 through 1994 were propargite, methyl bromide, trifluralin, simazine, and molinate (Gunier et al., 2001). On the other hand, the top pesticides used in California in 1994, based solely on weight, were sulfur, unclassified petroleum oil, methyl bromide, metam sodium, and copper hydroxide (CDPR, 2010). The differences between the top hazard-ranked and top applied pesticides by weight underscores how consideration of toxicity and exposure potential, in addition to pesticide use, can improve pesticide hazard prioritization in agricultural communities.

The current study applies a modified and expanded version of the Gunier et al. (2001) hazard-ranking scheme to prioritize pesticide hazards in Yuma County, Arizona from January 2006 through June 2011. It focuses specifically on Yuma County rather than the entire state of Arizona because pesticide use may vary spatially based on crops grown, pests targeted, and local climate (Aspelin and Grube, 1999). With improved specificity, it is possible to identify and prioritize community-specific pesticide hazards. In addition to ranking pesticide hazards for cancer, the scope has been widened with this study to include hazard-ranking schemes for endocrine disruption and reproductive/developmental toxicity, both notable health effects associated with chronic pesticide exposure. This study is one of the first to provide pesticide hazard-rankings for endocrine disruption and reproductive/ developmental toxicity. The most significant pesticide hazards in Yuma County, Arizona, with respect to cancer, endocrine disruption, reproductive/developmental toxicity, and overall chronic health risk are reported.

2. METHODS

2.1 Pesticide Use Data

Pesticide use in Yuma County for January 2006 to June 2011 was obtained from the Arizona Department of Agriculture Pesticide Application Database, referred to as the "1080

Database". The Arizona Department of Agriculture, Department of Environmental Quality (2011) requires all regulated growers or pesticide control advisors to submit a "1080" form for all pesticides applications containing an active ingredient listed on the Arizona Department of Environmental Quality Groundwater Protection List. The "1080" database includes application date, application location, crop to which the pesticide was applied, total acreage covered, chemical active ingredient, and chemical trade name. Only high-use pesticides in the hazard-ranking assessments that had a minimum annual average application of 1 pound/square mile of total land in Yuma County (5,519 pounds/year) were included (Gunier et al., 2001).

2.2 Determination of Associated Health Effects

Using a wide range of established criteria lists for cancer, endocrine disruption, and reproductive/developmental toxicity, each high-use pesticide was assessed for association with each health effect. The following cancer criteria lists were considered: Report on Carcinogens from the National Toxicology Program (NTP, 2011), Proposition 65 from the California EPA (CalEPA, 2011), Chemicals evaluated for carcinogenic potential: Corrected Annual Cancer Report from the USEPA (USEPA, 2010) and Agents classified by the IARC Monographs Volumes 1–104 from the International Agency for Research on Cancer (IARC, 2011).

For endocrine disruption, the criteria lists considered were the *Initial List of Chemicals for* Screening (USEPA, 2009) and *Second List of Chemicals for Screening* (USEPA, 2011b) from the USEPA Endocrine Disruptor Screening Program, Study on Enhancing the Endocrine Disruptor Priority List with a Focus on Low Production Volume Chemicals from the European Commission (Petersen et al., 2007), List of Potential Endocrine Disruptors from The Endocrine Disruption Exchange (Colborn et al., 1993), Endocrine Disruptors Strategy from the Illinois EPA (IEPA, 1997), Environmental Endocrine Disruptors: A Handbook of Property Data by Lawrence Keith (Keith, 1997), and Growing Doubt: A Primer on Pesticides Identified as Endocrine Disruptors and/or Reproductive Toxicants by Charles Benbrook (Benbrook, 1996).

For reproductive/developmental toxicity, the criteria list from Proposition 65 from the California EPA (CalEPA, 2011) and the USEPA Chronic (Non-Cancer) Toxicity Data for Chemicals Listed under the Emergency Planning and Community Right-to-Know Act, Section 313 (USEPA, 2000) were considered. See Supplementary Material p. 2–4 for more detailed information on each criteria list.

2.3 Hazard Factor Variables

Toxicity for each health effect and exposure potential based on chemical properties were used to calculate the hazard factor for each pesticide. Toxicity was assessed based on the following variables: "class" and "potency." Exposure potential was also based on two variables, "volatilization flux" and "soil persistence."

The class weight characterizes how strongly each pesticide is associated with each health effect based on the criteria lists on a scale of 1–10 from low to high. Consideration of multiple cancer criteria lists is an expansion of the Gunier et al. (2001) cancer "class" weighting system, which was limited to the cancer list provided by the USEPA. The cancer "class" weights in this study took into account the number of criteria lists in which the pesticide appeared and the level of the cancer-causing potential (e.g., USEPA "known" carcinogen versus "probable" carcinogen). See Supplementary Material, Table S1 for more detailed information on the cancer "class" weighting system. To assign a "class" weight for endocrine disruption, consideration was given to each criteria list's publication date and

whether it was based on peer-reviewed literature, in addition to the number of criteria lists in which the pesticide appeared and the reported level of endocrine disrupting potential. These extra considerations were given to compensate for the fact that endocrine disruption in humans is generally less understood than cancer, and its relationship with environmental exposures is more scientifically controversial (Daston et al., 2003; Fenner-Crisp et al., 2000). See Supplementary Material, Table S2 for the endocrine disruption "class" weighting system.

In the case that multiple class ratings could be applied, the higher rating was used because this is a more conservative assumption. Pesticides that did not appear on any of the criteria lists were given a class weight of 0 for that health effect, removing the pesticide from that hazard-ranking scheme. Formal "class" weights for reproductive/developmental toxicity were not applied because only 7 high-use pesticides were found on these criteria lists.

"Potency," the second variable of toxicity, was also weighed on a scale of 1–10 from low to high. The cancer slope factor was used to assign a cancer "potency" weight. For endocrine disruption and reproductive/developmental toxicity "potency" weights, the reference dose (RfD) was used. Cancer slope factors and RfD values were preferentially taken from the Integrated Risk Information System (IRIS) (USEPA, 1993; USEPA, 2011a). When information was not available through IRIS, other official USEPA documents, such as Registration Eligibility Decisions, were used to determine the cancer slope factor or reference dose (RfD). In the case that no cancer slope factor/RfD exists for the pesticide, a potency weight of "1" was applied. See the Supplementary Material for more information on the "potency" weighting systems.

To assess exposure potential of each pesticide, volatilization flux and soil persistence were both weighted on a scale of 1–10 from low to high. Volatilization flux is the pesticide's tendency to be transferred to the air from the ground. The "flux" weight is calculated using vapor pressure, soil adsorption, and aqueous solubility according to Glotfelty et al. (1989). The "persistence" weight was based on the pesticide's sandy-loam soil half-life because nearly 80% of Yuma, Arizona's soil is loam or sandy-loam (Holmes et al., 1905). See Supplementary Material, Table S3 for more information on how "potency," "flux," and "persistence" values were weighted.

2.4 Hazard Factor Calculation

Hazard factor values for each pesticide were calculated separately for cancer and endocrine disruption by multiplying the "class," "potency", "flux", and "persistence" weights and dividing by 1,000 to normalize hazard factors to values from 1 to 10 (Gunier et al., 2001). Since no "class" weight was assigned for reproductive/developmental toxicity, the hazard factor was computed as the product of the "potency," "flux," and "persistence" ratings divided by 100.

2.5 Hazard-Adjusted Pesticide Use

Hazard-adjusted pesticide use was computed by multiplying the computed hazard factors by the pesticide use (total weight applied) over the 4.5-year time period, as reported in the "1080" database provided by the Arizona Department of Agriculture. Pesticides were ranked for cancer, endocrine disruption, and reproductive/developmental toxicity according to their hazard-adjusted pesticide use for each health effect.

Pesticides were also ranked for overall chronic health effects, incorporating cancer, endocrine disruption and reproductive/developmental toxicity together. For the top 10 hazard-ranked pesticides for each health effect, the overall chronic health effects hazard for each pesticide was computed by dividing the "number of associated chronic health effects"

by the "average hazard-ranked position." The "number of associated chronic health effects" refers to how many of the chronic health effects in which the pesticide was ranked in the top 10. The "average hazard-ranked position" was computed by dividing the pesticide's placement on each specific chronic health effect endpoint hazard-ranking list by 3. The highest potential average hazard-ranking placement was 1.

3. RESULTS

3.1 Pesticide Use Data

From January 2006 to June 2011, 25.5 million pounds of active ingredients of pounds were applied in Yuma County, with an annual average application of 5.7 million pounds. A total of 223 different pesticide types were used throughout Yuma during this time period. After applying the minimum use criteria, 74 pesticides were considered high-use and thus, further assessed by the hazard-ranking schemes. Despite the relatively small number of pesticides evaluated, these 74 pesticides represented 98% of total applications, by weight, in Yuma County. The highest applied pesticides by weight were Aspergillus flavus 36 colonized wheat seed, bensulide, metam-sodium, trifluralin, and sethoxydim.

3.2 Pesticide Health Effects

Of the 74 pesticides considered for the hazard-rankings, 43 were associated with cancer, endocrine disruption, and/or reproductive/developmental toxicity (Table 1). Many of the pesticides were associated with more than one health effect. Approximately 80% of pesticide pounds applied in Yuma County during this time-period was associated with the chronic health effects of cancer, endocrine disruption, and/or reproductive/developmental toxicity.

3.3 Cancer Hazard-Ranking

The top 5 pesticides by cancer hazard factor out of the 21 pesticides associated with cancer were pronamide, maneb, metam-sodium, oxyfluorfen, and permethrin. The cancer hazard factors ranged across 2 orders of magnitude from 0.01–2.00 (Table 2). The hazard-ranking differed slightly once the pesticide use was adjusted by the cancer hazard factor. The top 5 pesticides by cancer hazard-adjusted use were metam-sodium, maneb, pronamide, mineral oil, and permethrin (Table 2).

3.4 Endocrine Disruption Hazard-Ranking

The top 5 pesticides by endocrine disruption hazard factor out of the 32 pesticides associated with endocrine disruption were trifluralin, pendimethalin, bensulide, endosulfan, and maneb. The endocrine disruption hazard factors ranged across 4 orders of magnitude from 0.004– 4.00 (Table 3). The top 5 pesticides by endocrine disruption hazard-adjusted use were trifluralin, bensulide, maneb, endosulfan, and chlorpyrifos (Table 3).

3.5 Reproductive/Developmental Toxicity Hazard-Ranking

There were only 7 pesticides associated with reproductive/developmental toxicity. The ranking of pesticides according to reproductive/developmental toxicity hazard factor were maneb; metam-sodium, bromoxynil octanoate, and EPTC (all ranked the same); bifenthrin; carbaryl; and streptomycin sesquisulfate. The hazard factors ranged two orders of magnitude from 0.05 to 2.00 (Table 4). After the hazard factor was used to adjust pesticide use, the ranking changed to maneb, metam-sodium, bifenthrin, carbaryl, and bromoxynil octanoate, EPTC, and streptomycin sequisulfate (Table 4).

3.6 Specific and Overall Chronic Health Effect Rankings

Taking into account all 3 chronic health effects together (i.e., cancer, endocrine disruption, reproductive/developmental toxicity), the top-ranked pesticides were maneb, metam sodium, trifluralin, pronamide and bifenthrin (Table 5).

4. DISCUSSION

The cancer hazard-ranking scheme developed by Gunier et al. (2001) was modified and expanded to prioritize pesticide hazards in Yuma County, Arizona. This study also developed and applied the first hazard-ranking schemes for endocrine disruption and reproductive/developmental toxicity that take into account pesticide use, toxicity, and exposure potential.

The relative order of hazard-ranked pesticides in this study was different from pesticide application by weight alone in Yuma County. The top hazard-ranked pesticides appear at the intersection of pesticide use, toxicity, and exposure potential. For example, pronamide was ranked the $4th$ highest overall pesticide hazard in Yuma, but it ranked $12th$ by pesticide application weight. In fact, pronamide's application was about 100,000 pounds/year less than maneb, the 4th highest applied pesticide by weight. Pronamide's high exposure potential via volatilization largely influenced pronamide's placement as the 4th highest overall hazard-ranked pesticide.

The hazard-ranking schemes effectively revealed pesticides of concern that might have gone unnoticed if pesticide application by weight was the only consideration. Most notably, pronamide and bifenthrin were the 4th and 5th highest overall hazard-ranked pesticides, despite the fact that neither pesticide appeared at all in the top 10 pesticides applied by weight for the same time period. The hazard-ranking schemes were similarly effective at deprioritizing pesticides with no associated health effects, even if they were highly applied. This was the case with Aspergillus flavus 36 colonized wheat seed, which was applied in a much greater quantity than any other pesticide, yet did not appear on any final hazardranking list because it was not included on the pre-established criteria lists for the considered health effects.

As expected, the hazard-adjusted use rankings in Yuma County, Arizona differed from the rankings for California by Gunier et al. (2001). Only trifluralin and metam-sodium appeared in the top 10 hazard-ranked pesticides in both studies. A partial reason for these dissimilarities is likely to be because of the unique pesticide use patterns in the two different study sites during two different study periods. In fact, some of the top hazard-ranked pesticides in Gunier et al.'s (2001) ranking, which considered pesticides used from 1991 through 1994, may no longer be used in this updated study period of 2006–2011. This is exemplified by methyl bromide, the second highest cancer hazard in the California study by Gunier et al. (2001), which has since been phased out by the USEPA due to evidence that it depletes stratospheric ozone (USEPA, 2011c), although a few exemptions continue to be granted. While methyl bromide, unsurprisingly, did not appear in the hazard-ranking for this current study, one of its most common substitutes, metam sodium, is currently the number one pesticide hazard for cancer and number two hazard for overall chronic health effects. Additionally, since the current study focused on one agricultural community within the state of Arizona, while Gunier et al. (2001) assessed pesticide hazards throughout the entire state of California, the captured resolution of each study is distinctive. While the Gunier et al. (2001) study was able to provide a general hazard ranking during that time period for California, it was not intended to capture hazards that are relevant to the many specific agricultural communities within California. The approach of the current study is novel

because it allows for agricultural communities to become informed specifically about the locally relevant hazards.

Carbaryl and permethrin are the only pesticides from the top 10 overall hazard-ranking from this study that were detected in Yuma's most recently published pesticide sampling study (CDC, 2002). Five of the current hazard-ranked pesticides in Yuma County (maneb, metam sodium, pronamide, bifenthrin, and mineral oil) were not analyzed in the Yuma sampling study, which sampled house and school dust from 1999 through 2000. While some of the hazard-ranked pesticides may not have been analyzed in the pesticide sampling study because of changes in pesticide use trends over the years, there may also have been a lack of appropriate identification of the most locally relevant pesticides for analysis.

Additionally, several of the pesticides that were detected in homes in the Yuma sampling study were not listed as the top hazards in the current study. This may largely be due to the fact that different pesticides are currently being used in Yuma compared to when the previous pesticide sampling was conducted. Other possible reasons for detection of certain pesticides in the home may be because the sampling occurred shortly after that particular pesticide had been applied in a nearby field, or because of weather conditions, such as a high wind event that promoted increased levels of that pesticide in the home. These details exhibit the importance of understanding intricacies such as temporal trends of pesticide use and environmental conditions in the agricultural community along with the hazard ranking schemes presented in this study.

An additional reason for detection of pesticides in homes that were not listed as the top hazards is the fact that considered pesticides were limited to those on the pre-established criteria lists for association with health effects. It is possible that some pesticide hazards were overlooked because they were not on the criteria lists. For example, although studies in the peer-review literature have found an association between diazinon exposure and cancer risk (Beane Freeman et al., 2005; Zhang et al., 2012) it did not appear on any of the cancer criteria lists, and thus was not considered as a carcinogen in this study. In the future, it would be useful to incorporate peer-reviewed literature to improve the toxicity component of the hazard rankings. Finally, while the current study considered pesticides applied during a 4.5 year time period, this may not capture pesticides applied in prior years that continue to persist in homes (Bradman et al., 2007; Curl et al., 2002). For instance, DDT was detected in homes in the Yuma sampling study (CDC, 2002), despite having been banned in 1972 (USEPA, 1972); however, DDT would not appear up on the hazard rankings because it is no longer legally applied.

Understanding chemical properties of pesticides that are relevant to exposure potential may provide insight regarding pesticide pathways into homes. For instance, maneb, the highest hazard-ranked pesticide for overall chronic health effects, binds strongly to soil (USEPA, 2005) suggesting that track-in and air-infiltration of wind-driven resuspended particles are both probable pathways into the home. Measuring maneb concentrations in the home during high and low winds can be used to assess the effect of wind-driven resuspension of particles. On the other hand, metam sodium is more likely to contaminate homes by air infiltration from pesticide spray drift, due to its high vapor pressure and volatilization flux (Lee et al., 2002; Li et al., 2006). High levels of metam sodium's toxic breakdown product, methyl isothiocyante (MITC), have been previously detected in air samples in an agricultural community (Merriman and Hebert, 2007). Metam sodium is thus a good candidate for future air sampling initiatives.

As research on the health effects of exposure to various pesticides accumulates, pesticide substitutions have become commonplace. In the early 2000s, organophosphate (OPs)

pesticides were widely used in Yuma County, as well as throughout the United States (Epstein and Bassein, 2003; Jaga and Dharmani, 2003). Chemical laboratories worked to improve analytical capabilities for OPs in response to the high demand for these services by researchers and regulatory agencies. Yet the current hazard-rankings reveal that only two of the top hazard-ranked pesticides, bensulide and chlorpyrifos, are members of the OP chemical class. As such, it is not currently reasonable to focus primarily on OPs in Yuma County. The hazard-ranking schemes provide a method to periodically reassess pesticide hazards in agricultural communities so that analysis for less relevant pesticides can be avoided.

This study is limited by self-reporting errors in the "1080" pesticide application database. Various spelling errors indicate that there may be low accountability for inputting thorough and accurate information. Identical records inputted consecutively in the database were observed on several occasions. It is unknown whether this is a duplication error or whether the pesticide was truly applied twice. In this study, each recorded application was assumed to be real and thus considered for the hazard rankings.

As mentioned earlier, this study is also limited to consideration of pesticides that appeared on the pre-established criteria lists and pesticides that were highly applied during the study period. A final limitation of this study is the fact that some pesticides are also approved for residential use, along with agricultural use, as is the case for both permethrin and carbaryl.

In this study, the relative ranking of pesticide hazards are each unique for cancer, endocrine disruption, and reproductive/developmental toxicity. This exhibits the value of considering more than one health effect when assessing overall risk for chronic health effects. It is important to remain current with the advances in research that have added the understanding of pesticide toxicity and associated health effects. The Gunier et al. (2001) study only considered cancer as a health outcome, but considerable advances have been made in understanding endocrine disruption and reproductive/developmental toxicity as an outcomes for pesticide exposure since that study period of 1991–1994. Therefore, it was natural to expand the hazard ranking scheme to incorporate endocrine disruption and reproductive/ developmental toxicity in the current study to more effectively assess health risk. Future studies could adapt and modify these hazard-ranking schemes to include other health effects, such a genotoxicity or neurodevelopment outcomes to continue this trajectory of improving assessments of health risks for agricultural communities.

5. CONCLUSIONS

Pesticide hazard-rankings schemes that consider exposure potential and association with various health effects, in addition to pesticide use, can help identify and prioritize pesticides of greatest health risk in agricultural communities. The highest hazard-ranked pesticides in this study were not necessarily the ones most applied by weight. Rather, the greatest pesticide hazards were occurred at the intersection of pesticide use, toxicity, and exposure potential. Mounting evidence of pesticide exposure being associated with a variety of health effects, along with the fact that the hazard-rankings were unique for each health effect, show the value of assessing pesticides for multiple health effects to better assess risk in agricultural communities. The current study has led the way in expanding hazard-ranking schemes to include endocrine disruption and reproductive/developmental toxicity, considering pesticide use, toxicity, and exposure potential. These pesticide hazard-ranking schemes can be applied in other agricultural communities as an effective tool to assess community-specific health risk for a variety of health effects.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Acronyms

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HIGLIGHTS

- **•** We apply pesticide hazard-ranking schemes for chronic health effects.
- **•** Our hazard-rankings consider pesticide use, toxicity, and exposure potential.
- We show that the highest hazard-ranked pesticides can vary by health effect.
- **•** These schemes can be applied to communities to identify local pesticide hazards.

Pesticides types and use in Yuma County (January 2006–June 2011) by health effect

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Overall ranking of pesticides for all chronic health effects

