Review

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State-of-the-art measurement of agricultural pesticide exposures

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Accurate exposure estimates are a common goal for risk assessment and epidemiologic research. Current methods for measuring agricultural pesticide exposure have not been fully validated. Epidemiologic studies use questionnaires to estimate exposures, producing job-exposure matrices and exposure algorithms. Biological monitoring can assist in the validation of questionnaire data. Pesticide risk assessments tend to rely on models derived from personal measurements of dermal exposure collected in quasi-experimental studies. Such studies often place constraints on exposure variability. Observational studies of occupational pesticide exposure that incorporate repeated measures are needed to improve the quality of exposure information. Pesticide exposure of family members can occur through spray drift and para-occupational exposures. Residential proximity to pesticide applications has been investigated recently with geographic information system (GIS), and global positioning system (GPS) technology. Biological monitoring can help to characterize exposure pathways in agricultural communities.

Key terms agricultural worker; agriculture; exposure assessment; pesticide; review.

Scientific methods for the measurement of pesticide exposures in agriculture have been developed to improve the accuracy of epidemiologic research and regulatory risk assessments. Advances have taken place in study designs, questionnaires, personal exposure monitoring, and biological monitoring to better characterize occupational pesticide exposures. New research has also focused on nonoccupational pesticide exposures in agricultural environments, such as exposures of families who live on or near farms and exposures of residents in agricultural communities. This paper discusses recent advances in these methods and highlights opportunities for new research in this field.

Occupational exposures

Study design and sampling strategies

Most epidemiologic studies of occupational pesticide exposure have case–control designs. Recent examples include a study of self-reported symptoms and the inhibition of acetylcholinesterase activity among Kenyan agricultural workers (1) and an evaluation of neuropsychological effects among French vineyard workers (2). Each of these studies demonstrated an association between self-reported symptoms or exposures and adverse outcomes, but neither was able to provide detailed documentation of exposure to specific agricultural chemicals. Prospective studies such as the Agricultural Health Study in the United States (3) offer an opportunity to develop such detailed exposure information. In the Agricultural Health Study persons with high pesticide exposure events have been identified (4), and specific pesticide-use patterns for these persons have been characterized (5). Recent efforts have focused on the development of a quantitative exposure algorithm for pesticide applicators in the study (6). These efforts should result in more accurate exposure assessments for people over extended time periods, increasing the possibility for testing etiologic relationships.

However, even these recent efforts lack actual measurements of personal exposure among study subjects. The exposure algorithm of the Agricultural Health Study is based on a combination of published studies and measurements contained in the Pesticide Handlers Exposure Database (PHED) (6).

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Teschke and her colleagues (7) have noted that the vast majority of personal measurement data for occupational pesticide exposures have been derived through quasi-experimental studies, and they have argued that the quasi-experimental study design can introduce bias into exposure measurements, since some requirements of these studies tend to constrain exposure variability (8). For example, such studies normally require strict compliance with all label instructions, the use of new personal protective equipment (PPE), and the use of recently serviced application equipment. While such work practices are clearly the goal of any program that emphasizes pesticide safety, they do not reflect the real-world conditions in most agricultural workplaces. Figure 1 illustrates, in hypothetical fashion, the impact of these constraints on exposure variability. The observational study results in a broader range of exposure values, whereas the quasi-experimental study reduces or eliminates high exposure events (eg, accidents or PPE failure) and reduces exposure for many study participants. In this example the geometric mean shifts from 32 to 22 exposure units. This inherent bias is one reason why van Hemmen (9) recommended that 90th percentile values be used rather than 50th percentile values as a more realistic descriptor of exposures in quasi-experimental studies.

If there is a need for more personal monitoring in observational studies of occupational pesticide exposure, it is also essential that such studies include repeated measures of exposure to account for within-worker (day-to-day) variability. A study by de Cock and his colleagues (10) found that, for reentry workers exposed to captan, within-worker geometric standard deviations were consistently higher than between-worker values. Simcox et al (11) reported similar findings for apple thinners exposed to azinphosmethyl. In summary, new studies of occupational pesticide exposure should be observational in nature, achieve probability-based sampling whenever possible, and include repeated measures of personal exposures. Incorporation of these study design features will greatly enhance our understanding of exposures in this worker population.

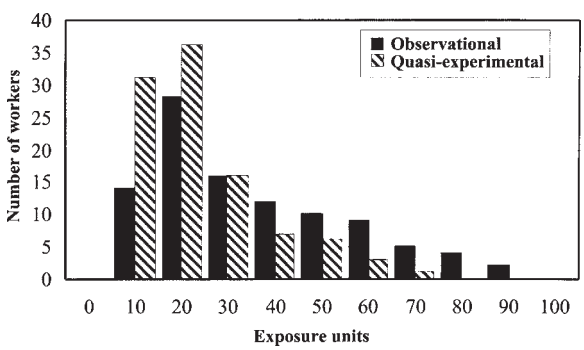

Figure 1. Distribution of two sets of 100 hypothetical personal-exposure measurements from pesticide applicators derived from observational and quasi-experimental scenarios. The quasi-experimental scenario reflects truncation of the top 10% of the exposure values, reducing values to 70 or less; it also reflects a downward shift of 25% of the values to simulate exposure constraints during such studies. Geometric means and standard deviations for the two scenarios are 32 (SD 1.9) and 22 (SD 1.8) for the observational and quasi-experimental scenarios, respectively.

Personal monitoring of dermal exposure

The measurement of dermal exposure to pesticides has proved to be a complex problem within the field of exposure assessment. Techniques for the measurement of pesticide exposure to the skin fall into the following three general categories, each of which has been reviewed recently: surrogate skin techniques (12), chemical removal techniques (13), and fluorescent tracer techniques (14, 15).

Surrogate skin techniques. Surrogate skin techniques include patches and whole-body garments. The patch technique relies on an assumption of exposure uniformity within regions. If exposure patterns are not uniform, then patches may overestimate or underestimate exposure. Despite this limitation, patch data have proved useful for characterizing differences in exposures due to work activities, application procedures, engineering controls, and chemical protective clothing use. Researchers in British Columbia recently used the patch data found in the PHED to develop a job-exposure matrix for agricultural workers (16). The quantitative exposure algorithm proposed for the Agricultural Health Study also employs patch data from the published literature in conjunction with the PHED data for many of the algorithm inputs (6).

Limitations inherent in patch sampling can be overcome by sampling entire anatomical regions with garments. Whole-body dosimeters have been proposed as a standard method for measuring pesticide handler exposures (17). This method has proved cumbersome and expensive and, therefore, has not been used widely. The major assumption underlying this approach is that the collection medium captures and retains chemicals in a manner similar to that of skin. Concerns include overestimates of exposure if the garment retains more chemical than skin and underestimates if breakthrough occurs. None of the garment samplers in current use has been systematically tested for retention efficiency. Thus the accuracy of whole-body dosimeters remains an open question.

Chemical removal techniques. Chemicals deposited on skin can be removed through washing or wiping. Handwash sampling procedures can normally be standardized to ensure that they are operator-independent so that studies can be compared, whereas skin wiping relies on

procedures that are inherently operator-dependent and thus include an unknown component of variability. Measurements of chemical removal represent only what can be removed from the skin at the time of sampling rather than at the actual skin loading, but some studies have considered wash values as "exposure", relying on an implicit assumption that removal was 100%. Fenske & Lu (18) reported that only 50% of the pesticide chlorpyrifos was removed when skin was washed immediately after exposure, and less than 25% was removed 1-hour postexposure. Furthermore, removal efficiency decreased with decreased skin loading. These findings demonstrate the need to conduct appropriate efficiency studies of laboratory removal prior to the use of hand washing in field studies. A recent study by Marquat et al (19) included both a laboratory and field component for several pesticides. Removal efficiency in the field was consistently lower than corresponding values in the laboratory. These findings suggest that even efficiency studies of laboratory removal may not provide adequate adjustment factors for the interpretation of exposure measurements in the field.

Skin wiping has found increasing use in recent years, but, to date, no standard protocol for this technique has been established by governmental regulatory agencies or international bodies such as the World Health Organization (WHO) or the Organization for Economic Cooperation and Development (OECD). Table 1 provides data from recent laboratory studies of skin wiping efficiency (13). In most cases only about half of the deposited material was removed from the skin. The only study that did not indicate a significant underestimation with this technique was that of Geno et al (20). This experiment can be viewed as a kind of "best-case" study, since the residence time of the chemical on the skin was essentially zero. It is also striking that, for both chlorpyrifos and pyrethrin I, the sampling efficiency values increased across the three sampling days. In the case of pyrethrin I, for example, for days 1, 2, and 3, the average removal efficiencies were 64%, 89%, and 123%, respectively. While the average removal efficiency was 92 (SD 27)% for the 12 trials, the daily removal efficiencies significantly differed from each other [$P<0.001$ in an analysis of variance (ANOVA)]. This marked trend of increasing removal efficiency suggests that pyrethrin I may have remained on the hands of the participants between trials (the same persons repeated the study on consecutive days). If one assumes that the pyrethrin I that was not removed by handwiping was available for removal in the subsequent trial, then the average removal efficiency drops to 72 (SD 13)%, and the differences between the daily means are not significant (ANOVA $P=0.06$).

Field studies that have evaluated the accuracy of skin-wipe sampling are limited. A study of apple thinners exposed to azinphosmethyl compared glove, handwash, and handwipe measurements after a 2-hour exposure period (21). As illustrated in figure 2, skinwipes removed only 15% of the amount removed by handwashing. Skinwipe measurements were estimated to be approximately 10% of the true exposure. These results indicate that the skinwipe sample values collected in field studies will require substantial adjustment if they are to be used as dermal exposure measurements.

Fluorescent tracer techniques. Dermal exposure can be quantified directly and noninvasively through the use of fluorescent whitening agents and video-imaging analysis (22). The technique has proved useful in the estimation of dermal exposures among golf course workers (23), greenhouse applicators (24, 25), and other agricultural workers (26).

Ideally, this method can provide improved accuracy over other methods, since it measures actual skin-loading levels, requires no assumptions regarding exposure distribution over skin surfaces, and can identify hitherto

Table 1. Laboratory studies of skin-wipe sampling efficiency.[a] (residence time = time between application and skin wipe)

Compound	Species	Residence time[b] (hours)	Percentage removed from skin
Propoxur	Human	4	48
Glyphosate	Pig	1.5	42
Methyl parathion	Pig	1.5	47
Alachlor	Pig	1.5	54
Trifluralin	Pig	1.5	57
Pyrethrin I	Human	0[b]	92
Chlorpyrifos	Human	0	104

[a] Data adapted from table 2, Brower et al (13).
[b] Removal occurred immediately following skin contact.

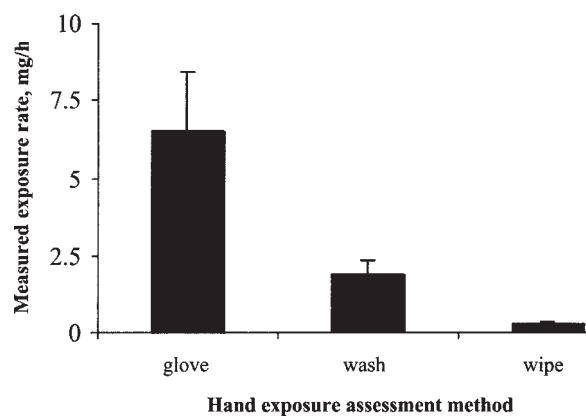

Figure 2. Comparison of three dermal-exposure assessment methods in a group of apple thinners exposed to azinphosmethyl (mean exposure rates in milligrams per hour, plus standard deviation). The wipe sample measurements were 23 times lower than the glove measurements and 6.5 times lower than the hand-wash measurements. Adapted from Fenske et al (21).

unrecognized exposure pathways. In practice, however, the demonstration of measurement accuracy has proved problematic. The most successful validation study of this technique demonstrated a strong association between the deposition of fluorescent whitening agents on the skin and biological measurements of primicarb exposure in greenhouse workers (24).

Biological monitoring

Urine and saliva sampling. Pesticides or their metabolic products have been measured in the urine of workers for some 40 years. The key challenges associated with this technique are interpretive rather than analytical, as very low concentrations of these chemicals can now be measured with reasonable accuracy and precision. Chester (17) proposed that controlled pharmacokinetic studies on human volunteers should be a prerequisite for the use of biological exposure data in risk assessments. Woolen (27) proposed several conditions for the development of a valid biological monitoring method. First, animal studies should be used to design human studies. Second, human studies should conform to the Helsinki Declaration for Conduct of Human Volunteer Studies. Third, urinary excretion of the unchanged pesticide or metabolite should account for at least 30% of the orally administered dose. Fourth, the range between persons should not exceed a factor of three. These criteria provide a good starting point for biological monitoring studies. In any such studies, the uncertainties associated with extrapolation from measurements in urine to internal dose need to be identified explicitly. Extrapolation of human doses from animal pharmacokinetic data is not desirable, as the uncertainty associated with the resulting dose estimates cannot be determined.

Saliva has been explored as a practical medium for monitoring exposures to several environmental chemicals, including pesticides. The feasibility of saliva biomonitoring for atrazine exposure has been studied in a systematic manner using an animal model (28). In these studies, salivary concentrations of atrazine were found to be highly correlated with plasma concentrations under varying conditions, as illustrated in figure 3. A study of herbicide applicators demonstrated that saliva sampling is feasible under realistic work conditions (29). Figure 4 indicates that atrazine concentrations in saliva were highest at the end of the workshift, the concentrations decreasing in bedtime and next morning samples.

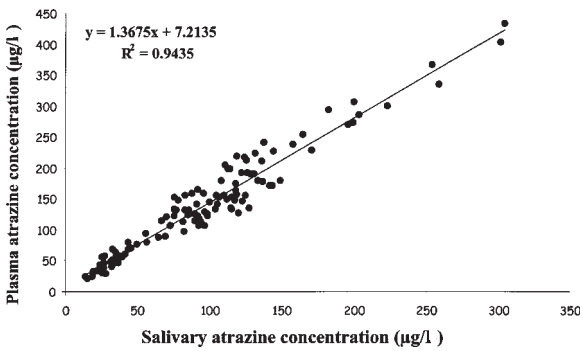

Figure 3. Correspondence of saliva and plasma atrazine concentrations in 18 rats following intravenous administration (N=124). Adapted from Lu et al (28).

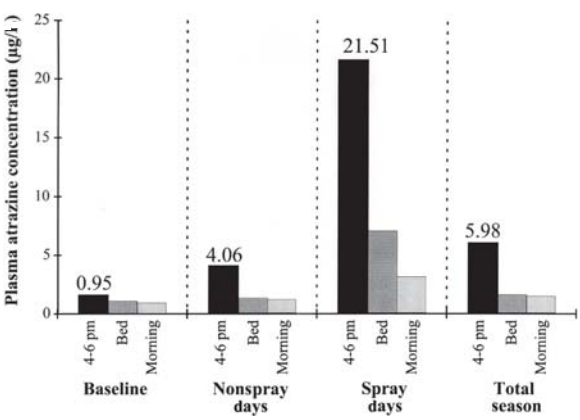

Figure 4. Median salivary atrazine concentrations in a group of herbicide applicators at three time points: 4–6 pm, bedtime, and next morning. The measurements reported are at baseline, for nonspray days, for spray days, and for the total season. Adapted from Denovan et al (29).

Validation studies with biological monitoring. Several recent studies have employed urinary measurements of the herbicide 2,4-dichlorophenoxyacetic acid (2,4-D) to evaluate the validity of questionnaire data. In these studies the biological measurements were assumed to be accurate predictors of internal dose. One study used 2,4-D concentrations measured in 24-hour urine samples from 126 applicators to examine self-reported exposure factors (30). Pesticide formulation, protective clothing and gear, application equipment, handling practices, and personal hygiene practices were all significant predictor variables, and a multivariate model explained 39% of the total variance (R^2=0.39). A second study involved 98 professional turf applicators (31). In this case, 24-hour urine samples were collected for several days, and weekly 2,4-D doses were modeled from these data. The amount sprayed, type of nozzle used, and use of gloves were significant predictors of exposure, and a multivariate model explained 63% of the total variance (R^2=0.63). It seems likely that the detailed dose estimates developed in this study improved the accuracy of the model. The use of biological monitoring to evaluate self-reported exposure information is a promising approach for epidemiologic research. The thoughtful design of both these studies provides an instructive template for researchers in this field.

Residential and community exposures

Farm family exposures

Children and spouses of farmers and agricultural workers may be exposed to agricultural chemicals through their daily activities beyond exposures associated with diet. Parents may bring pesticides home from work on skin, clothing, or workboots. Such para-occupational or take-home exposure has been observed for many occupations (32). Several studies have included farm family members as a part of occupational exposure assessments (10, 33, 34).

Studies in Washington State have used house dust and urine sampling of preschool children to investigate this pathway, documenting a difference across occupational and nonoccupational groups (35, 36). A more recent study found a strong association between pesticide concentrations in dust samples from commuter vehicles and residences (37).

Studies of residential proximity to pesticide-treated farmland

Community exposures can also result from spray drift resulting from agricultural pesticide applications. A recent study sampled 44 preschool children in an agricultural community for 18 months, collecting urine on a bi-weekly basis (38). Pesticide metabolite concentrations in urine increased during periods of active spraying and returned to normal levels when spraying ended. This pattern is illustrated for the dimethyl dialkylphosphate metabolites in figure 5.

Residential proximity to agricultural spraying has been the focus of several recent epidemiologic investigations. Researchers in California have taken advantage of that state's Pesticide Use Reporting Database to classify populations according to pesticide-use patterns near residences. For example, a study of fetal deaths due to congenital abnormalities considered total pesticide usage within an area of 9 square miles (1 mile = 1.6 kilometers) (39). A more narrow definition of exposure reduced the area to the 1 square mile in which the residence was located. A recent study of childhood leukemia in California used the same database, but also used geographic information system (GIS) methods to assign exposure values on the basis of census blocks (40). This approach provided a more-refined exposure estimate based on the specific location of the residence relative to spraying in the surrounding region.

GIS technology has now been used in several studies to provide more accurate information regarding residential proximity to farmland. A recent study used satellite images and historical farm records to identify specific crop locations and the locations or residences relative to these crops (41). The United States Environmental Protection Agency conducted a pilot study on the impact of residential proximity to agricultural fields on metabolite concentrations of an organophosphorus pesticide in the urine of toddlers (42). While residential proximity to agricultural fields can provide useful information regarding exposure potential, knowledge of children's behavior in relation to fields and to agricultural spraying is also needed. A recent study has addressed this concern through the development of a data-logging global positioning system (GPS) unit that can be worn by young children (43). This technology provides time-specific location data for the child throughout the day. When this information is linked to pesticide use information and field measurements of air concentrations and pesticide deposition on surfaces in a child's activity zone, more precise estimates of exposure can be obtained.

In summary, the exposure of farm family members, family members of agricultural workers, and those who live in agricultural communities represents a new area

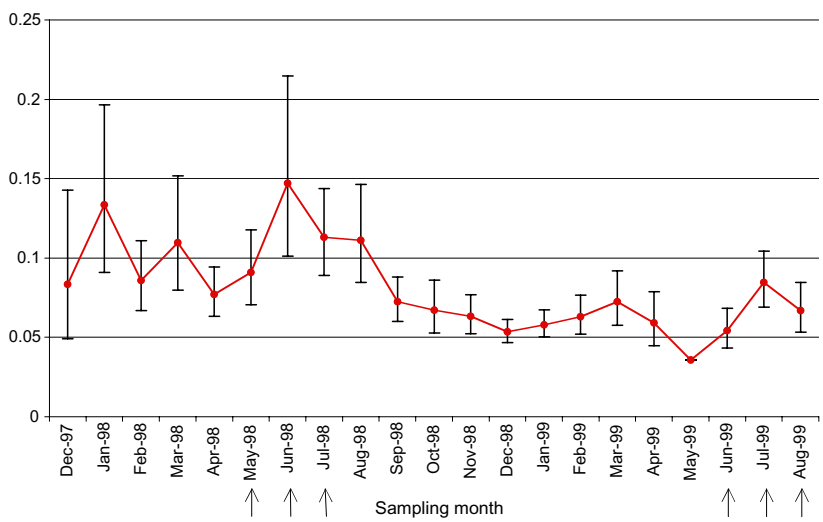

Figure 5. Dimethyl dialkylphosphate (DAP) metabolite concentrations of organophosphorus pesticides in the urine of preschool children living in a tree fruit region of Washington State. Geometric means (μmol/l) and 95% confidence intervals are provided for each month. Arrows indicate months of dimethyl organophosphorus pesticide spraying in the region. Adapted from Koch et al (38).

of research and a challenge for the field of exposure assessment. The multi-route, multi-pathway nature of such exposures will require more advanced study designs and statistical analysis, as well as more accurate exposure measurement tools.

Concluding remarks

Current exposure measurement techniques for dermal exposure to pesticides are limited in their accuracy and precision. New research should be directed towards improving these techniques. Biological monitoring should be included as a complementary method in such exposure assessments. Biological monitoring measurements in field studies should be preceded by human pharmacokinetic studies. Biological monitoring can also be used effectively to evaluate questionnaire data collected in the course of epidemiologic research. The assessment of exposures among farmer and agricultural worker family members and in agricultural communities is a challenging field that has employed new technologies to quantify important exposure factors such as residential proximity to agricultural fields and children's activity patterns. New efforts to refine such methods are warranted for populations with high exposure potential.

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