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Dietary intakes of pesticides based on community duplicate diet samples

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HIGHLIGHTS

- Intakes were successfully obtained using a new community based sampling technique.
- Community dietary exposure may differ from national database information.
- Exposure to both OP and pyrethroid pesticides were consistent with a dietary model.
- Pesticide usage patterns change which should be considered in future studies.
- Community Duplicate Diet Method is a tool for researchers to meet emerging needs.

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ABSTRACT

The calculation of dietary intake of selected pesticides was accomplished using food samples collected from individual representatives of a defined demographic community using a community duplicate diet approach. A community of nine participants was identified in Apopka, FL from which intake assessments of organophosphate (OP) and pyrethroid pesticides were made. From these nine participants, sixty-seven individual samples were collected and subsequently analyzed by gas chromatography/mass spectrometry. Measured concentrations were used to estimate dietary intakes for individuals and for the community. Individual intakes of total OP and pyrethroid pesticides ranged from 6.7 to 996 ng and 1.2 to 16,000 ng, respectively. The community intake was 256 ng for OPs and 3430 ng for pyrethroid pesticides. The most commonly detected pesticide was permethrin, but the highest overall intake was of bifenthrin followed by esfenvalerate. These data indicate that the community in Apopka, FL, as represented by the nine individuals, was potentially exposed to both OP and pyrethroid pesticides at levels consistent with a dietary model and other field studies in which standard duplicate diet samples were collected. Higher levels of pyrethroid pesticides were measured than OPs, which is consistent with decreased usage of OPs. The diversity of pyrethroid pesticides detected in food samples was greater than expected. Continually changing pesticide usage patterns need to be considered when determining analytes of interest for large scale epidemiology studies. The Community Duplicate Diet Methodology is a tool for researchers to meet emerging exposure measurement needs that will lead to more accurate assessments of intake which may enhance decisions for chemical regulation. Successfully determining the intake of pesticides through the dietary route will allow for accurate assessments of pesticide exposures to a community of individuals, thereby significantly enhancing the research benefit realized from epidemiological exposure studies.

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Abbreviations: CTEPP-NC, Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants Study in North Carolina; JAX, Biological and Environmental Monitoring for Organophosphate and Pyrethroid Pesticide Exposure in children Living in Jacksonville, FL; MNCPES, Minnesota Children's Pesticide Exposure Study; NCS, National Children's Study; NHANES, National Health and Nutrition Examination Survey; OP, Organophosphate; PDP, Pesticide Data Program; RPD, relative percent difference; SHEDS, Stochastic Human Exposure And Dose System; TDS, Total Diet Study; USDA, United States Department of Agriculture; USDHHS, United States Department of Health and Human Services; USEPA, United States Environmental Protection Agency; WWEIA, What We Eat In America.

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1. Introduction

Collection of samples in epidemiological exposure studies requires protocols that will optimize research resources and minimize burden to study participants. While collection and analysis of environmental samples can be one of the most costly portions of a study, these costs are offset by the substantial increase in the accuracy of the critical information gained on potential exposures and routes. Among all environmental samples, the diet sample is often ignored entirely for this reason and because of its relatively high collection burden on participants. Alternatively, dietary exposures may be estimated from questionnaire responses (USEPA, 2010), but potential contamination from the residential environment is missed. Since diet has been shown to provide the highest potential for exposure to pesticides (Morgan et al., 2007; Hu et al., 2004; Lu et al., 2004), an accurate determination of dietary pesticide exposure necessitates collection and analysis of food samples.

A commonly-used approach for estimation of dietary exposure has involved the collection of duplicates of individual consumed foods, whereby participants provide duplicate portions of all foods eaten over the monitoring period (Melnyk et al., 1997; Bradman et al., 2007; Riederer et al., 2010). Excessive sample analysis cost and high participant burden prohibit use of this approach in studies where thousands of participants are enrolled, such as the National Children's Study (NCS). The Community Duplicate Diet Methodology provides a protocol that allows food samples to be collected so as to minimize the use of resources (Jordan et al., 2010; Melnyk et al., 2012). Additionally, as the community of interest becomes larger, the proportion of participants needed in the study to achieve a representative sample will decrease due to the constant sample size needed to test specific hypotheses.

National databases of food consumption and pesticide residues are valuable sources of information in estimating dietary exposure, but may lack the ability to select foods based on defined demographic and cultural groups and also lack the important pesticide contribution associated with food storage and preparation in the residence, (U.S. Department of Agriculture et al., 2006a; U.S. Department of Agriculture et al., 2006b; U.S. Department of Health and Human Services et al., 2007a; U.S. Department of Health and Human Services et al., 2007b). The community duplicate diet approach used information in these databases to inform sample collection schemes while relying on the increased accuracy and representativeness of collecting actual food samples from individual's homes. This unique technique takes advantage of existing database information together with needed collection of foods to obtain a better estimate of dietary exposure for a community, an important assessment requirement for determining linkages between environmental stressors and potential health outcomes.

Participant responses to questionnaires administered during the Apopka field study indicated that the community duplicate diet approach was feasible as a dietary collection strategy (Melnyk et al., 2012). The true test of the utility of this approach would be in the determination of intake of selected pesticides. The estimation of dietary intake was accomplished on samples collected from members of a defined demographic community which allowed for an accurate assessment of the potential exposures to individuals within this community.

2. Methods and materials

A demographically well-defined community was identified for an observational field study conducted in Apopka, FL to assess the feasibility of a new tool that could provide adequate food samples to measure dietary intake on a large scale (Melnyk et al., 2012). This was an "observational research" study, as defined in 40 CFR Part 26.402. The study protocol and procedures to obtain the informed consent of participants were reviewed and approved by an independent institutional review board and they complied with all applicable requirements of the Common Rule. Assumptions included; 1) the individuals in the nine-person community were reasonable representatives of a larger, demographically-similar community of Hispanic women of child-bearing age, 2) probability of potential dietary exposure risk was similar across the representatives, 3) food consumption and pesticide residue information obtained from demographically-similar records in the targeted national databases was applicable to the study community, 4) any seemingly unusual findings were actually typical for the community, and 5) the environment in which the community resides provided a potential source for pesticide contamination of the food. Following the newly developed food collection and analysis protocols (Melnyk et al., 2012), the necessary samples were obtained to determine dietary exposure to pesticides.

Food consumption and pesticide residue databases were used to guide participant recruitment in order to provide a study community with relatively similar dietary behaviors and to increase the chances for measurable pyrethroid levels in collected samples. The top 20 most commonly prepared and eaten foods for Hispanic women of child-bearing age were determined based on information gathered from extant databases. This information was obtained from published food frequency questionnaire data contained in national surveys and was summarized as percentages of each food relative to all foods consumed by Hispanic women of child-bearing age. The National Health and Nutrition Examination Survey (NHANES), What We Eat In America (WWEIA), USDA's Pesticide Data Program (PDP), and FDA's Total Diet Study (TDS) were used to develop the list of foods frequently consumed by members of the study population (U.S. Department of Agriculture et al., 2006a; U.S. Department of Agriculture et al., 2006b; U.S. Department of Health and Human Services et al., 2007a; U.S. Department of Health and Human Services et al., 2007b). Foods encompassed all meals eaten within a day (i.e., breakfast, lunch, dinner and snacks). Using this information, nine women in Apopka, FL were recruited and asked to collect their commonly consumed prepared foods. From our *a priori* use of the databases, these foods were expected to have a finite probability of containing permethrin pesticide residues. Participants were included in the study based on the foods they normally consumed; they were in no way encouraged or influenced to change their dietary behaviors in order to participate. The collection of the food samples and the associated questionnaire data were performed by field technicians deployed to the field site; both the technical assessment of the collection process and the participant's responses to the questionnaires and food diaries were used to determine feasibility of collection, acceptability of instructions and tools, and overall burden to participants. Details of the protocol implementation and individual pesticide analyses of 67 food samples collected during the observational study are summarized in Melnyk et al., 2012.

Each of the nine participants provided up to eight individual prepared food samples segregated by eating events, i.e., breakfast, lunch, dinner, and snack. While the most frequently consumed individual foods served as the basis for participant selection, the actual foods collected from the participants were prepared foods, containing the individual components described below, as presented "at the table." In this case, an individual sample was defined as a composite of foods as consumed, i.e., "salad" consisting of lettuce, carrots, etc. or "rice and beans" consisting of rice, beans, and onions. Only one eating event was included in a single sample bag. The contents of the bags were reviewed by the field technician against the information listed in the food diaries to ensure compliance with the instructions, correct food identification, and to estimate the proportions of individual meal food items in each bag.

Food samples were analyzed by gas chromatography/mass spectrometry (GC/MS) using a single method for six organophosphate (OP) (MDLs ranging from 0.05 to 0.1 ng/g) and eleven pyrethroid pesticides (MDLs ranging from 0.05 to 0.8 ng/g). Foods were homogenized, extracted, concentrated, and subjected to solid phase extraction prior to analysis by GC/MS in the selected ion monitoring mode (Melnyk et al., 2012). In addition to the measured pesticide residues, consumption information was obtained from the food diary collected during