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Correlation of pesticide exposure from dietary intake and bio-monitoring: The different sex and socio-economic study of children

Sukesh Narayan Sinha*, Venkat Reddy Banda

Food and Drug Toxicology Division, National Institute of Nutrition (ICMR), Jamia-Osmania P.O., Hyderabad – 500 007, AP, India

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ABSTRACT

Exposure to organophosphorus pesticides was assessed though bio-monitoring of urinary dialkyl phosphate metabolites to characterize children's exposure to pesticides. No reports have been found which evaluate exposure of pesticides among adolescents of different sexes. The primary objective was to develop a coupled exposure–dose modeling approach that can be used to determine the metabolite concentrations. Related objective was to determine any significant changes of pesticide exposure among the adolescents of different sexes. The primary objective was to develop a coupled exposure-dose modeling approach to determine the metabolites concentrations to keep children's urine metabolites levels below specified values considering exposures from water, and the food related objective was to determine any significant changes of pesticide exposure among the adolescents of different sexes. We recruited a sample of 377 children (188 boys, 189 girls) ages 6–10 and 11–15 years from Hyderabad, India for urine collection. Results showed that the mean concentrations of dialkyl phosphate metabolites in first morning first urine samples (3.05 μmol L^{-1}) were strongly correlated with concentrations of the same-day 24-h samples (1.7 μmol L^{-1}) (r = 0.997, model $R^2 \approx 0.994$, $p < 0.00$) with 99.4% accuracy. Irrespective of similar amounts of conventional food consumption, girls showed 87.5% of detection frequency of DAP metabolites which was higher than the 74% detection frequency of DAP metabolites among boys. The female group showed (87%) higher pesticide metabolite levels than boys. This report may help to focus on new studies of the connection between adolescents of different sex and organophosphorus metabolite exposure and to develop an exposure database to facilitate health risk assessment in our day-to-day environment.

1. Introduction

Telangana and Andhra Pradesh account for a hefty 24% share of pesticide consumption in the country. These two states are grappling with increased pesticide residues in food commodities. The central government program launched by Indian council of agriculture research, Ministry of Agriculture, Government of India, named "Monitoring of Pesticide Residues at National Level" conducted by the Government of India in 2014–15 showed that 2.6% of all samples of commodities contained pesticide residues (chlorpyrifos, acephate, fenitrothion) above the Maximum Residues Limits. During the program, National Institute of Plant Health Management (NIPHM), Hyderabad, India reported the concentration of organophosphorus pesticides (fenitrothion, endosulfan, chlorpyrifos, acephate, monocrotophos, and acetamiprid) in vegetable samples. It denotes that the organophosphate (OP) pesticides were among the major pesticides being used around Hyderabad" (MPRNL, 2014).

OP pesticides are well known to metabolize to dialkylphosphates metabolites (DAP), which are excreted in urine (Lambert et al., 2005). Determination of these metabolites is useful for assessing human exposure to organophosphates. Several reports have shown that OP pesticides were the first class of pesticides for which tolerances were reassessed because of their common mode of toxicity, widespread use, and unknown long-term health effects (Wesseling et al., 2002; EPA, 2003). Therefore, in many epidemiologic studies, markers of exposure in biologic samples have been measured to estimate the absorbed dose. One of the most common ways to assess OP pesticide dose is by quantifying six common urinary DAP metabolites (Lu et al., 2006; Lambert et al., 2005; Bradman et al., 2007; Bouchard et al., 2010; Zheng et al., 2011; Oulhote and Maryse, 2013). Several studies have reported that spot sample analysis collection will provide basis for the exposure assessment of the population to OP pesticides (James and Roberts, 2012; Berenbaum and Snyder, 1995).

Children are uniquely vulnerable to uptake and adverse effects of pesticides because of developmental, dietary and physiologic factors (Goldman, 1993; Jacqueline et al., 2004; Landrigan et al., 1999;

* Correspondence to: Food and Drug Toxicology, National Institute of Nutrition (ICMR), Jamia-Osmania P.O., Hyderabad 500007, Telangana, India.
E-mail address: sukeshnr_sinha@yahoo.com (S.N. Sinha).

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Tanner, 1971). Exposure occurs though ingestion, inhalation, or dermal contact. Unintentional ingestion by children may be at considerably higher doses because of greater intake of food or fluids per pound of body weight compared to adults. In particular, the potential effects of pesticide exposure to the developing fetus and children are of interest to society and regulatory agencies (Rauh et al., 2011). Although, the neurotoxic associations of high-level prenatal and early childhood exposure to certain pesticides are well established, the implications of potential effects observed at low exposures are less straightforward, particularly in the absence of a clinically defined adverse outcome. Studies evaluating potential neurodevelopment effects associated with pesticide exposure are challenging to interpret, in part because of the diversity of types and classes of chemicals, differences in exposure measures, and the wide range of instruments used to assess outcomes (Slotkin and Seidler, 2012; Rauh et al., 2011, 2006; Eskenazi et al., 2007). Nevertheless, it is important to critically evaluate the evidence to date, as well as to identify important research gaps and methodological issues that require further attention to advance the understanding of observed effects.

In many parts of the world, the usage of OP pesticides started way back in 1960s several studies were conducted on the toxicity of OP pesticides along with the usage of OP pesticides and it is still going (Akgür et al., 2003; Eskenazi et al., 1999; Freed et al., 1976; Cortés-Eslava et al., 2015; Bondarenko et al., 2004; Dhanushka and Peiris, 2017; Vale, 1998). In developed countries, especially in US, where several bio-monitoring reports are available concerning the exposure of children of different age groups to OP pesticides (Bradman et al., 2007, 2013; Kissel et al., 2005; Lu et al., 2010). India to date, has not reported on exposure assessment of OP pesticides in children of different sexes with the same dietary habit, environment, body weight, mean age. Therefore, the study was designed to assess the exposure of OP pesticides among children in the South Indian city of Hyderabad.

2. Methods

2.1. Study population

The Scientific Advisory Committee and the International Ethical Committee of the National Institute of Nutrition, India had approved the study. The study included 377 children residing in Hyderabad city. Out of them, 188 were boys and 189 were girls, belonging to either 6–10 or 11–15 year age groups. The children were in apparently healthy with no history of diabetes or renal disease, toilet-trained, free of involuntary urination, especially at night, and had local language (Telugu or Hindi) speaking mothers, who were at least 24 years old. Further, children who were consuming any form of organic food, apart from the conventional diet were excluded from the study. All information concerning diet and eating habits were obtained from their parents. The mothers were explained about the study and asked for their consent to enroll their children for the study. Enrollment was limited to one child per household and written informed consent was obtained from their parents. As per an earlier study by Sinha et al., 2012a, the Hyderabad area was divided into five zones (viz.; south, north, east, west and central). In each zone, farmers' vegetable market and local vegetable street outlets were selected. A sample of 500 g of each vegetable variety was collected from farmers' vegetable market as well as vegetable street outlets in each zone. Ten grams of the samples from each vegetable sample collected from market and street outlet was analyzed by LC–MS/MS. Urine samples were also collected from these zones and it was confirmed that in each household they were purchasing fruits and vegetables in their respective markets as well as from street outlets (Sinha et al., 2012a, 2012b). Recently, a report released by the Ministry of Agriculture Government of India on concentration of pesticides on vegetable crops and prevalence of OP pesticides indicates that the use of OP pesticides around Hyderabad region is considerably high (MPRNL, 2014).

2.2. Interviews

Parents were interviewed during the first visit and information was collected on the child's age, weight, parents' age, occupation, annual family income, home ownership, the length of stay at the current residence, and housekeeping practices. Further, information on the use of pesticides in and around the home, on the home structure, in the garden, on the lawn, and on the pets was collected from the parents. And, further questions regarding how long it had been since the most recent application of pesticides in each of these areas, whether the applicator had been someone from the home or not. They were requested to inform about any applied pesticide products, if available at home at the time of interview in order to record the product name, Indian pesticide Registration number date and location of applicant.

2.3. Food diaries

In our study, the "thee cups method" was used to measure the quantities of food consumed by children. The three cups were C1, C2, and C3. The first cup, C1 (volume was 200 ml) was used to measure the quantities of cooked foods such as *plain rice*, *lemon rice*, *fried rice*, and *upma*. The second cup, C2 (volume was 150 ml) was used to measure cooked vegetables such as eggplant, cabbage, cauliflower, tomato and okra and fruits such as grapes. And, the third cup, C3 (volume was 100 ml) was used to measure the liquid food items such as *fruit juices*, *milk*, and *beverages*. Parents were provided with these measuring cups and instructed to maintain a food diary. They were asked to note down the food consumed by the children for two days before and on the day of collecting urine samples. They were also directed to include the foods consumed outside home (e.g., food consumed during lunch at the school) in the food diary. They recorded the types and the approximate amounts of all conventional foods consumed by their children for breakfast, lunch, and dinner. After all the food items were recorded, they were collected and converted into “units of servings” for each child.

2.4. Urine collection

On the first visit, the study staff had provided the sampling supplies to the parents, which included sampling record forms, 500-ml polypropylene bottles, gloves, and large collection bottles with blank labels. They were instructed to collect the first urine samples of their wards early in the morning and the remaining voids. The collected urine bottles were kept in plastic container and stored in the refrigerator, until retrieval on the following day. Parents identified and separated each sample bottle as an FMV (First-morning void) or a non-FMV spot sample, and even noted down the time of the sample collection. Research staff reviewed the sampling records to assure accuracy and completeness. The samples were then collected, and transported in dry ice to the laboratory at the National Institute of Nutrition and were processed immediately. The transport time was maximum 30 min. The total volume of urine of each child was measured and stored at −20 °C until the analysis. The 24 h samples included 6–10 collected voids (the mean was 7.8). Of all the first-morning urine samples collected, 95.8% (range, 50%100%) reported voids and 63% of the 24-h samples were based on 100% collection of all voids. Reasons for missed voids were out-of-wash room, toileting accidents, and participant's errors. After adjustment for missed voids, the average volume, collected per child, was 817 ml and ranged from 150 ml to 1584 ml.

2.5. Samples processing and analysis

Samples were, processed at the National Institute of Nutrition, Hyderabad, India. The weight (in grams) and volume (in milliliters) of each void was measured (Salita et al., 1998). Individual voids, from 24-h sampling sessions, were not selected for individual analysis, except the first-morning void which was later compared with the pooled sample. After that, the samples were aliquoted, and stored at −20 °C until the analysis. On the day of analysis, 5 ml of each sample was taken and lyophilized,