Environment International 92–93 (2016) 294–300

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Overall and class-specific scores of pesticide residues from fruits and vegetables as a tool to rank intake of pesticide residues in United States: A validation study

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ARTICLE INFO

Article history:
Received 3 February 2016
Received in revised form 21 March 2016
Accepted 16 April 2016
Available online xxxx

Keywords:
Fruits/vegetables pesticide residues
Organochlorine pesticide
Organophosphate pesticide
Validation study
USDA Pesticide Data Program
NHANES

ABSTRACT

Pesticide residues in fruits and vegetables are among the primary sources of pesticide exposure through diet, but the lack of adequate measurements hinder the research on health effects of pesticide residues. Pesticide Residue Burden Score (PRBS) for estimating overall dietary pesticide intake, organochlorine pesticide score (OC-PRBS) and organophosphate pesticide score (OP-PRBS) for estimating organochlorine and organophosphate pesticides-specific intake, respectively, were derived using U.S. Department of Agriculture Pesticide Data Program data and National Health and Nutrition Examination Survey (NHANES) food frequency questionnaire data. We evaluated the performance of these scores by validating the scores against pesticide metabolites measured in urine or serum among 3,679 participants in NHANES using generalized linear regression. The PRBS was positively associated with a score summarizing the ranks of all pesticide metabolites in a linear fashion (p for linear trend <0.001). Furthermore, individuals in the top quintile of this score had urinary pesticide metabolite levels 13.0% (95% CI 8.3%–17.7%) higher than individuals in the lowest quintile. Similarly, we observed significant associations of the OC-PRBS and OP-PRBS with the levels of lipid-adjusted total serum organochlorine pesticides and urinary creatinine-adjusted organophosphate pesticides, respectively. The relative difference (RD) in average pesticide metabolite rank between extreme quintiles was 17.8% (95% CI: 11.1%–24.4%, p for trend <0.001) for the OP-PRBS, whereas the RD was marginally significant at 7.0% (95% CI: −0.5%–14.4%, p for trend 0.07) for the OC-PRBS. The PRBS and OP-PRBS had similar performance when they were derived from fruits and vegetables with high vs. low pesticide residues, respectively (p for trend <0.001 for all associations). The OP-PRBS was associated with all measured organophosphate pesticides, whereas the positive association between OC-PRBS and averaged measured organochlorine pesticide residue rank was primarily driven by hexachlorobenzene. OC-PRBS had better performance when derived from more contaminated fruits and vegetables (p for trend 0.07) than from less contaminated Fruits and vegetables (p for trend 0.63), although neither of the associations achieved statistical significance. The PRBS and the class-specific scores for two major types of pesticides were significantly associated with pesticide biomarkers. These scores can reasonably rank study participants by their pesticide residue exposures from fruits and vegetables in large-scale environmental epidemiological studies.

1. Background

A pesticide is defined as any substance or mixture of substances intended for preventing, destroying, repelling, or mitigating any pests (insects, mites, nematodes, weeds, rats, and etc.). Based on their targeted pests, they can be categorized into insecticides, herbicides, fungicides, and various other substances (EPA, 2009). Based on their chemical structure, three main classes of pesticides are carbamates, organophosphates and organochlorinated pesticides. These pesticides are widely applied to fruit and vegetable crops worldwide, and herbicides are mostly used for maize in the developed countries (Zhang et al., 2011). Pesticide Monitoring Program 2012 report by U.S. Food and Drug Administration (USDA) showed that a considerably higher proportions of domestic fruits and vegetables had detectable pesticide residue or had residue levels exceeding the EPA standard, in comparison to other foods, including dairy, grain products and fish (FDA, 2012),

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http://dx.doi.org/10.1016/j.envint.2016.04.028
0160-4120/

suggesting that intake of fruits and vegetables is among the major sources of pesticide residue exposure.

Dietary guidelines have consistently suggested increasing the consumption of fruits and vegetables for the prevention of major chronic diseases (U.S. Department of Health and Human Services and U.S. Department of Agriculture, 2015) based on unequivocal evidence from clinical trials and epidemiological studies (Bazzano et al., 2003, 2002; Hu and Willett, 2002; Joshipura et al., 2001; Krauss et al., 2000; Liu et al., 2000; Mozaffarian et al., 2003; Muraki et al., 2013; Rimm et al., 1996; U.S. Department of Health and Human Services and U.S. Department of Agriculture, 2015). Meanwhile, concerns have been raised regarding increased exposures to pesticide residues resulting from higher consumption of these healthful foods (Bhanti and Taneja, 2007; Boobis et al., 2008). One study specifically evaluated pyrethroid exposure through food consumption and concluded that part of the variation in pyrethrois intake is explained by vegetable intake but not other food sources (Fortes et al., 2013). Data from the USDA Pesticide Data Program (PDP) have revealed that pesticide residues were common for a variety of fruits and vegetables (USDA, 2015a). In Europe and China, pesticide residues have also been identified in multiple fruits and vegetables, and the residues of certain pesticides were found to be more than maximum residue level values recommended by European Union, World Health Organization and Food and Agricultural Organization (Bakırcı et al., 2014; Yu et al., 2016). The concerns regarding the impacts of food-borne pesticides on human health have led advocacy organizations to develop an index to rank food items in terms of pesticide residue levels based on the PDP data (EWG, 2014).

Indeed, several short-term intervention studies have showed that young children's exposure to organophosphate pesticides, the most widely used pesticides in agriculture sectors (Chen et al., 2009; Sharma et al., 2010; Wang et al., 2008), is primarily from diet (Bradman et al., 2015; Lu et al., 2008, 2006). In addition, although some organochlorine pesticides such as hexachlorobenzene have been banned from use in U.S. for 50 years, their residues persist in soil and water because of their long half-lives and thus they may still affect human health (Jones and de Voogt, 1999). Actually, a few organochlorine pesticides are still in use in U.S., such as 2,4 dichlorophenoxyacetic acid (2,4-D). However, the long-term health effects of exposures to pesticide residues through fruits and vegetables consumption remain to be elucidated.

Table 1
Demographical information of NHANES participants (2003–2004) by quintile of the overall fruits and vegetable pesticide score (PRBS).[a]

Demographical information	Overall fruits and vegetable pesticide score (PRBS), quintiles				
	1	2	3	4	5
N	392	392	392	392	392
Age, years					
<20	50.8	43.6	39.0	31.4	34.4
20–40	22.2	20.4	19.1	20.2	16.1
40–60	12.5	14.8	18.6	19.1	18.1
≥60	14.5	21.2	23.2	29.3	31.4
Gender, male	54.6	53.8	46.2	40.1	40.3
White race	46.2	49.5	48.5	49.7	37.8
Body mass index, kg/m^2					
<25	55.8	51.8	48.7	40.8	49.7
25–30	23.5	26.5	26.0	29.9	28.6
≥25	20.7	21.7	25.3	29.3	21.7
Smoking status					
Never smoker	74.5	72.5	68.9	71.7	74.7
Past smoker	12.5	14.8	18.9	20.7	17.4
Current 1–10 cigarettes/day	6.1	5.1	5.6	3.3	4.1
Current 11–20 cigarettes/day	5.1	5.1	5.1	3.8	3.6
Current 21 or more cigarettes/day	1.8	2.6	1.5	0.5	0.3
Vigorous or moderate activity over past 30 days	56.4	60.5	54.6	61.2	57.9
Pest control in past month	20.4	18.9	17.9	15.8	18.9

[a] Data are proportion (%).

Quantifying concentrations of pesticide metabolites in human biospecimens to measure pesticide exposures is ideal but challenging. High costs, the lack of reliable assays for some pesticides, and short half-lives of pesticide metabolites poses major challenges for the evaluations of pesticides' health effects in large-scale epidemiological studies. In contrast, pesticide residue scores may be more cost-effective and can be a useful tool to facilitate the preliminary examination of pesticide residues in relation to human health before investigations based on biomarkers are implemented. Chiu et al. recently described a novel pesticide score to summarize the overall exposure to pesticide residues from fruits and vegetables and evaluated its association with semen quality (Chiu et al., 2016, 2015). Results suggested that the pesticide score, but not fruits and vegetables per se, were associated with poor semen quality. In the current study, we aimed to improve Chiu's pesticide score algorithm by combining the fruit and vegetable consumption frequency and their respective pesticides residues levels to form a Pesticide Residue Burden Score (PRBS) that can reflect the overall pesticide exposures attributed to fruit and vegetable intake. We further derived two class-specific (organochlorine and organophosphate) pesticide scores because pesticides in classes may have heterogeneous effects on human health. We subsequently examined the associations of the overall score and the two class-specific scores with a variety of pesticides measured in urine or serum in the 2003–2004 National Health and Nutrition Examination Survey (NHANES).

2. Material and methods

2.1. USDA Pesticide Data Program (PDP)

In 1991, the USDA PDP was launched to estimate the potential risk of pesticide residues in food (USDA, 2015b). About 15 types of fresh and processed fruits and vegetables have been randomly sampled and measured for pesticide residues each year since 1992, and the sampling procedures were designed to capture actual residues in the food supply as close as possible to the time of consumption. The fruits and vegetables sampled varied over time. In addition, fresh samples were washed or peeled before testing to emulate the practices of the average consumers. The PDP database contains information for over 300 types of environmental pesticides found in fruits and vegetables. In the current study, we used 2003–2006 PDP data to develop the pesticide scores to match the time when pesticide metabolites were measured in 2003–2004 NHANES. A total of 29 fruits and vegetables that were presented in both NHANES food frequency questionnaire and PDP data were considered in the current analysis. Eighteen types of fruits and vegetables were measured in 2003–2004 PDP programs. We used 2005–2006 PDP data, which were based on eleven types of Fruits and vegetables, to supplement 2003–2004 PDP data in order to include more fruits and vegetables in the current analysis.

2.2. Development of Pesticide Residue Burden Score (PRBS)

The primary goal of the PRBS is to rank study participants in terms of their overall exposures to pesticide residues from fruit and vegetable intake as previously described (Chiu et al., 2015). Specifically, in the USDA PDP database, we used three indexes to estimate an overall pesticide residue profile for each fruit or vegetable: 1) the percentage of samples with any detected pesticides; 2) the percentage of samples tested with pesticides exceeding the tolerance level; and 3) the percentage of samples with three or more types of pesticides detectable (USDA, 2015c). For each of these three indexes, fruits and vegetables were ranked into tertiles and were assigned a score ranging from 1 (bottom tertile) to 3 (top tertile). Then scores based on these three indexes are summed for each fruit or vegetable to create a pesticide rank score. The possible range of pesticide rank score is from 3 (least contaminated) to 9 (most contaminated). For example, strawberries are in the top tertiles for all three indexes and therefore have a pesticide rank score of 9, whereas onions have a pesticide rank score of 3 because they are in the bottom