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Associations between dietary factors and urinary concentrations of organophosphate and pyrethroid metabolites in a Canadian general population

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ABSTRACT

Objectives: Few studies to date have characterized the relationships between diet and urinary concentrations of pesticides. In the current study, associations between dietary factors and urinary concentrations of organophosphate and pyrethroid metabolites were examined in a Canadian general population using data from the Canadian Health Measures Survey (CHMS).
Methods: In the CHMS, urinary concentrations of dialkylphosphate (DAP) and pyrethroid metabolites were measured in 5604 participants aged 6–79 years. Associations between dietary factors and total concentrations of DAP (ΣDAP) and pyrethroid metabolites (ΣPYR) were examined.
Results: Over 90% of participants had at least one type of DAP and 99.8% had pyrethroid metabolites detectable in urine samples. After adjusting for age, sex, race/ethnicity, immigrant status and body mass index, ΣDAP among participants with high (3rd tertile) fruit consumption was 1.43 (95% CI: 1.26–1.61) times the concentration among those with low (1st tertile) consumption. ΣDAP was also positively associated with vegetable consumption, for those with high consumption ΣDAP being 1.33 times (95% CI: 1.16–1.52) the concentration for those with low consumption. ΣPYR among participants with high vegetable consumption was 1.42 (95% CI: 1.23–1.66) times the concentration among those with low vegetable consumption. ΣPYR was also positively associated with pulses/nuts consumption (p-values < 0.01) and household use of chemicals to control head lice or pet flea ($p = 0.002$).
Conclusions: Positive associations between dietary factors and urinary concentrations of organophosphate and pyrethroid metabolites suggest greater regulation of pesticide use on food products may help to reduce pesticide exposures and associated health risks among the general population.

Introduction

Organophosphate (OP) and pyrethroid (PYR) pesticides are currently widely used in large quantities in agriculture and residential areas to control insect pests (FAO, 2006). According to the Environmental Protection Agency in the US, only 1% of the five billion pounds of pesticides applied annually actually reaches its target (EPA, 2011), raising the possibility of human expose to OP and pyrethroid pesticides through the environment.

Pesticide residues in food products are potentially a major source of pesticide exposure to humans. It has been estimated that approximately 33% of table-ready food items, including wheat-based product, fruit and fruit juice, have detectable levels of one or more types of pesticides, especially post-harvest insecticides such as chlorpyrifos (Nougadere et al., 2012). A study in Denmark showed that pesticide residues were detected in 60% of fruits and 18% of vegetables sold (Poulsen and Andersen, 2003). Studies from the USA found that the OP pesticide, chlorpyrifos and its degradation products, were detected in up to 97% of unspecified solid food samples (MacIntosh et al., 2001; Morgan et al., 2005). Results from the Children's Pesticide Exposure Study (CPES), conducted in Seattle (USA) and Atlanta (USA), showed that OP and pyrethroid insecticides were respectively detected in 14% and 5% of children's nonorganic fruit, vegetable and fruit juice samples (Lu et al., 2010). In addition, cypermethrin, a synthetic pyrethroid pesticide, was detected in more than 30% of the composite diet samples collected from an adult population in the USA, including dairy, fruits, grains, beans/nuts/legumes and both above- and below-ground vegetables

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(Riederer et al., 2010). According to the U.S. Department of Agriculture Pesticide Data Program (PDP), 36% of food items sold at the market contained at least one type of pesticide or pesticide metabolites (Pesticide Data Program Annual Summary Reports, 2013).

Association studies have suggested relationships between dietary intake and urinary concentrations of OP and PYR insecticides (Berman et al., 2013; Fortes et al., 2013; Lu et al., 2009; McKelvey et al., 2013; Munoz-Quezada et al., 2012; Riederer et al., 2008). In an adult population in Israel, urinary concentrations of OP metabolites among subjects with high fruit consumption (>75th percentile) were 1.27 times ($p = 0.073$) of the concentrations among those with low fruit consumption (<75th percentile) (Berman et al., 2013). Results from the New York City Health and Nutrition Examination Survey (NYC HANES) showed that urinary OP concentrations were positively associated with increasing frequency of fruit consumption, with adjusted mean urinary concentrations of OP metabolites being 141 nmol/L (95% CI: 102, 195 nmol/L) for subjects consuming fruit more than once per day, and 74 nmol/L (95% CI: 59, 94 nmol/L) for those who ate less than two servings per week. (McKelvey et al., 2013). Another study in Chile also showed that residual levels of OP pesticides on fruits were significantly associated with higher urinary concentrations of OP metabolites among school-aged children (Munoz-Quezada et al., 2012). A recent study of an adult population in Italy showed that consuming cruciferous (OR: 4.67; 95% CI: 1.07–20.5) and leafy vegetables (OR: 6.88; 95% CI: 1.50–31.7) was strongly associated with high levels (≥0.70 μg/g creatinine) of pyrethroid metabolite 3-phenoxybenzoic acid (3-PBA) in urine (Fortes et al., 2013). In the US National Health and Nutrition Examination Survey (NHANES, 1999–2002), dietary factors, including eating spinach, lettuce and broccoli and drinking citrus juices, were significantly associated with urinary concentrations of 3-PBA for children, adolescents and adults (Riederer et al., 2008). Moreover, results from the US CPES showed that there was an approximate 50% reduction in urinary concentrations of 3-PBA when substituting conventional diet with organic food among children, which suggests that dietary intake was an important predictor of urinary 3-PBA levels (Lu et al., 2009). However, these exposure studies were typically conducted in specific age groups (Berman et al., 2013; Lu et al., 2009; Munoz-Quezada et al., 2012), and often in a small convenient sample (Fortes et al., 2013) or specific region (McKelvey et al., 2013). Only few large general population-based studies (McKelvey et al., 2013; Riederer et al., 2008), and none in Canada, have investigated the relationship between dietary factors and urinary concentrations of OP and PYR metabolites.

In the current study, associations between dietary factors and urinary concentrations of organophosphate and pyrethroid pesticide metabolites were examined in a Canadian general population using the data from the Canadian Health Measures Survey (CHMS) Cycle 1. During the CHMS-Cycle 1 sampling period (2007–2009), there were 15 organophosphates and 10 pyrethroids registered for use in Canada (Supplementary Table S1 and S2) (Health Canada, 2010) and among these, the organophosphate insecticides azinphos-methyl, chlorpyrifos, diazinon, phorate, phosalone, phosmet and terbufos, were restricted to agricultural use (Health Canada, 2009).

Methods

In 2007–2009, Statistics Canada conducted the Canadian Health Measures Survey (CHMS-Cycle 1), a population-based cross-sectional survey, to collect baseline health information of Canadians (Statistics Canada, 2011). In the current study, we used the data on 5604 participants aged 6–79 years in the CHMS-Cycle 1.

A multi-stage sampling strategy was used by Statistics Canada to select CHMS-Cycle1 participants: collection sites were selected after being stratified by geographic region and CMA (Census Metropolitan Area); dwellings within each collection site were sampled and then stratified based on inhabitants' ages; in each age stratum, participants were then sampled from the selected dwellings (Statistics Canada, 2011). People living on reserves and Aboriginal settlements, residents of institutions, members of the Canadian Forces and those living in remote areas with population density less than 400 people per square kilometer, were not included (Statistics Canada, 2011). Detailed information on the CHMS-Cycle 1 can be obtained from the online publication of Statistics Canada (Statistics Canada, 2011).

In the current study, urinary concentrations of OP and pyrethroid pesticide metabolites, and information on demographic, anthropometric, socioeconomic, household pesticide use and dietary food consumption of 5604 subjects were obtained from the CHMS-Cycle 1. For children 6–11 years of age, parents or guardians provided information for the household questionnaire. On behalf of children aged 6–13, parents or guardians also gave consent to participate the clinical examination and completed the screening questions in the clinical questionnaire during the clinic visit (Statistics Canada, 2011).

Urinary concentrations of OP and pyrethroid pesticide metabolites

Approximately 60 mL of mid-stream urine was collected for each CHMS-Cycle 1 participant aged 6–79 years (Statistics Canada, 2011). After collection, urine samples were refrigerated immediately and transported to an analytical laboratory at the National Public Health Institute of Quebec (INSPQ). Urine samples were analyzed for dialkyl phosphate (DAP) metabolites, including dimethyl phosphate (DMP), dimethyl thiophosphate (DMTP), dimethyl dithiophosphate (DMDTP), diethyl phosphate (DEP), diethyl thiophosphate (DETP) and diethyl dithiophosphate (DEDTP), and for metabolites of pyrethroid pesticides (Mikata et al., 2012), including *cis*-DCCA [*cis*-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane-1-carboxylic acid], *trans*-DCCA, *cis*-DBCA [*cis*-3-(2,2-dibromovinyl)-2,2-dimethylcyclopropane-1-carboxylic acid], 3-PBA (3-phenoxybenzoic acid) and 4-fluoro-3-PBA (Statistics Canada, 2011). Concentrations of DAPs and PYR metabolites were determined by gas chromatography-mass spectrometry (GC-MS) (Health Canada, 2010; Statistics Canada, 2011). Limits of Detection (LOD) for measuring DMP, DMTP, DMDTP, DEP, DETP, DEDTP, 3-PBA, 4-fluoro-3-PBA, *cis*-DCCA, *trans*-DCCA and *cis*-DBCA were 1.0 μg/L, 0.6 μg/L, 0.3 μg/L, 1.0 μg/L, 0.6 μg/L, 0.3 μg/L, 0.01 μg/L, 0.008 μg/L, 0.007 μg/L, 0.01 μg/L and 0.006 μg/L, respectively (Health Canada, 2010). Urinary creatinine concentration (g/L) was measured using the colorimetric Jaffe method and was used to normalize pesticide metabolite concentrations for urine dilution (Barr et al., 2005).

Demographic, anthropometric and socioeconomic factors

Information on age, sex, race/ethnicity, immigration status and province of residence were collected using the CHMS-Cycle 1 questionnaire (Statistics Canada, 2010). Standing height and weight were measured using standard procedure of the Canadian Physical Activity, Fitness and Lifestyle Approach (CPAFLA) (CSEP, 2003). Body mass index (BMI) was calculated using the formula weight (kg)/[height (m)]^2 (CSEP, 2003). Subjects were classified, according to the US Center for Disease Control and Prevention (CDC) BMI-for-age growth charts for children aged 2–19 years (Kuczmarski et al., 2002) and the WHO International Classification of BMI for adults aged 20 years and above (WHO, 2004), into three BMI groups: underweight or normal [BMI <85th percentile for children and