



Urinary concentrations of organophosphate and carbamate pesticides in residents of a vegetarian community

T. Berman^{a,*}, T. Göen^b, L. Novack^c, L. Beacher^c, L. Grinshpan^d, D. Segev^e, K. Tordjman^f

^a Public Health Services, Ministry of Health, Israel

^b Institute and Outpatient Clinic of Occupational, Social and Environmental Medicine, Friedrich-Alexander-University of Erlangen-Nuremberg, Erlangen, Germany

^c Department of Public Health, Faculty of Health Sciences, Ben Gurion University of the Negev, Beer Sheva, Israel

^d Robert H. Smith Faculty of Agriculture, Food and Environment, Hebrew University, Rehovot, Israel

^e Braude College, Karmiel, Israel

^f Institute of Endocrinology, Metabolism and Hypertension, Tel Aviv Sourasky Medical Center, Tel Aviv, Israel

ARTICLE INFO

Article history:

Received 30 May 2016

Received in revised form 28 August 2016

Accepted 28 August 2016

Available online 31 August 2016

Keywords:

Vegetarian

Pesticides

Exposure

Biomonitoring

Organophosphates

Dialkylphosphates

ABSTRACT

Few population studies have measured urinary levels of pesticides in individuals with vegan, vegetarian, or organic diets. The objectives of this study were to evaluate whether a vegan/vegetarian diet was associated with increased exposure to organophosphate and carbamate pesticides, and to evaluate the impact of organic consumption on pesticide exposure in vegans and vegetarians. In the current pilot study conducted in 2013–2014, we collected spot urine samples and detailed 24 h recall dietary data in 42 adult residents of Amirim, a vegetarian community in Northern Israel. We measured urinary levels of non-specific organophosphate pesticide metabolites (dialkylphosphates, (DAPs)) and specific metabolites of the current-use pesticides chlorpyrifos (3,5,6-trichloro-2-pyridinol (TCPy)), propoxur (-isopropoxyphenol (IPPX)), and carbaryl (1-naphthol). Six DAP metabolites were detected in between 67 and 100% of urine samples, with highest geometric mean concentrations for dimethylphosphate (19.2 µg/g). Creatinine-adjusted median concentrations of total DAPs and of TCPy were significantly higher in Amirim residents compared to the general Jewish population in Israel (0.29 µmol/g compared to 0.16, $p < 0.05$ for DAPs and 4.32 µg/g compared to 2.34 µg/g, $p < 0.05$ for TCPy). Within Amirim residents, we observed a positive association between vegetable intake and urinary TCPy levels ($\rho = 0.47$, $p < 0.05$) and lower median total dimethyl phosphate levels in individuals reporting that >25% of the produce they consume is organic (0.065 µmol/L compared to 0.22, $p < 0.05$). Results from this pilot study indicate relatively high levels of urinary organophosphate pesticide metabolite concentrations in residents of a vegetarian community, a positive association between vegetable intake and urinary levels of a chlorpyrifos specific metabolite, and lower levels of total dimethyl phosphate in individuals reporting higher intake of organic produce. Results suggest that consumption of organic produce may offer some protection from increased exposure to organophosphate pesticide residues in vegetarians.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Residues in food are the dominant source of pesticide exposure in the general population (McKone et al., 2007). In Israel, both organophosphate (OP) and carbamate pesticides are widely used in agricultural crops, with 232 tons of OP pesticides and 55 tons of carbamate pesticides sold for agricultural use in 2010 (Israel Bureau of Statistics, 2014). Pesticide residue data from 2013 to 2014 indicates that between

4 and 8% of sampled agricultural products on the market in Israel have pesticide residue levels above the maximum residue level, and that residues of the OP pesticides chlorpyrifos, azinphos-methyl, dichlorvos and methamidophos, and the carbamate pesticides methomyl, oxamyl, and carbofuran were detected in sampled food items (Israel Ministry of Health, 2013; Israel Ministry of Health, 2014).

Both organophosphate and carbamate pesticides are rapidly metabolized and excreted from the body in hours to days, with biological half-lives between 12 and 36 h (Barr, 2008; California Environmental Protection Agency, 1997). As metabolites are excreted in urine, exposure to OP and carbamate pesticides can be measured by analyzing urinary concentrations of compound-specific metabolites such as the chlorpyrifos metabolite 3,5,6-trichloro-2-pyridinol (TCPy), the propoxur metabolite 2-isopropoxyphenol (IPPX), the carbaryl metabolite 1-naphthol, and by measuring non-specific OP metabolites

Abbreviations: OP, Organophosphates; DAPs, Dialkylphosphates; TCPy, 3,5,6-trichloro-2-pyridinol; IPPX, 2-isopropoxyphenol; DMP, Dimethylphosphate; DMTP, Dimethylthiophosphate; DMDTP, Dimethyldithiophosphate; DEP, Diethylphosphate; DETP, Diethylthiophosphate; DMDTP, Diethyldithiophosphate.

* Corresponding author at: Department of Environmental Health, Public Health Services, Ministry of Health, Jeremiya Street 39, 9446724 Jerusalem, Israel.

E-mail address: tamar.berman@moh.health.gov.il (T. Berman).

(dialkylphosphates (DAPs)). OP and carbamate metabolites have been measured in several human biomonitoring studies in adult populations, for example in the National Health and Nutrition Examination Survey (DAPs, OP and carbamate-specific metabolites), the Canadian Health Measures Survey (DAPs), the French adult population (DAPs), a study of Danish mothers (DAPs), and in several longitudinal birth cohort studies (DAPs and TCPy) (Centers for Disease Control and Prevention, 2015; Report on Human Biomonitoring of Environmental Chemicals in Canada, 2010; Fréry et al., 2010; Harley et al., 2016; Danish Environmental Agency, 2015).

Previous studies have documented associations between consumption of fruits or vegetables and OP pesticide exposure in both adults and children in populations in the US (McKelvey et al., 2013), Canada (Ye et al., 2015), Chile (Muñoz-Quezada et al., 2012), Spain (Roca et al., 2014), and Israel (Berman et al., 2013b). Consistent with findings on the association between fruit and vegetable consumption and OP exposure, organic diets have been shown to lead to a substantial reduction in OP exposure in both children and adults. Studies have shown significant decreases in several OP pesticide metabolites in urine in rural, urban, and suburban children during several days of eating organic food (Bradman et al., 2015; Lu et al., 2006). In Australian adults, mean total OP pesticide metabolite levels were 89% lower during the one-week organic diet phase compared to the conventional diet phase (Oates et al., 2014). In addition, in a study on long term dietary exposure to OPs, individuals who reported eating organic food at least occasionally – when matched on produce intake – had lower urinary DAP levels than those who ate primarily conventional food (Curl et al., 2015).

While the association between diet and exposure to OP pesticides is well documented, there have been few studies on pesticide exposure in vegans, vegetarians and none on the association between organic consumption and pesticide exposure in this sub-population. In a study on pesticide residue dietary intake in the French general population and the vegetarian population, vegetarians were preferentially exposed to pesticides for which fruit, vegetables and cereals were indicated as main contributors, including the OPs chlorpyrifos-methyl and diazinon (Van Audenhaege et al., 2009).

We previously reported that exposure to OP pesticides in the general adult population in Israel is high relative to the US and Canada, possibly due to relatively high consumption of fresh, regionally produced fruits and vegetables (Berman et al., 2013a; Berman et al., 2013b). In the current study, we measured urinary concentrations of OP and carbamate metabolites in residents of Amirim, a cooperative community in northern Israel based on vegetarian, vegan, and organic lifestyle. Amirim residents are expected to follow communal rules, including not to smoke in public places, to maintain a vegetarian or vegan diet, and to refrain from using pesticides (Amirim website, 2016). The aims of the study were to evaluate whether a vegan/vegetarian diet was associated with higher exposure to OP and carbamate pesticides and to evaluate the impact of organic consumption on pesticide exposure in this population.

2. Materials and methods

2.1. Recruitment

We recruited a convenience sample of 42 adult residents of the Amirim community in Northern Israel between December 2013 and June 2014. The study protocol was reviewed and approved by the Tel Aviv Sourasky Medical Center Helsinki Committee. Written informed consent was obtained for all respondents. All analysis of data for the study was conducted without details on the identity of the participants.

2.2. Questionnaire

Each participant filled in a detailed 61-item health, lifestyle and dietary questionnaire, including personal medical history and medication intake. The questionnaire included specific questions on recent use of

household pest control and *anti-flea* pet products. Participants then responded to a 24 h dietary recall based on the Israel National Health and Nutrition Survey. For each of the food items consumed, participants were specifically asked if the item or food ingredients were organic. In order to generate a semi-quantitative score for organic product consumption, we determined the proportion of organic food items consumed by dividing the number of organic food items by the total number of food items consumed in the past 24 h. Proportion of organic food consumption was stratified into 4 categories: <10%; between 10 and 25%, between 25 and 50% and over 50%.

2.3. Urine sample collection

Spot urine samples were collected in BPA and phthalate-free 100 mL polypropylene urine containers (Sarstedt, Nümbrecht, Germany) at the time of interview. All urine samples were maintained at 4 °C for a maximum of 8 h until they were transported to Tel Aviv Sourasky Medical Center. Urine samples were aliquoted and frozen at –80 °C. Within 6 months of collection, urine samples were shipped to the University of Erlangen – Nuremberg in Germany on dry ice (–70 °C), where they were analyzed.

2.4. Sample analysis

OP and carbamate exposure was assessed by analyzing urinary concentrations of TCPy (chlorpyrifos and chlorpyrifos-methyl metabolite), IPPX (propoxur metabolite), 1-naphthol (carbaryl metabolite) and the non-specific OP parameters dimethylphosphate (DMP), dimethylthiophosphate (DMTP), dimethyldithiophosphate (DMDTP), diethylphosphate (DEP), diethylthiophosphate (DETP) and diethyldithiophosphate (DMDTP) in urine. Additionally, 2-naphthol in urine was analyzed for the differentiation of carbaryl and naphthalene exposure.

Laboratory analyses of pesticide metabolites and creatinine were performed at the Institute and Outpatient Clinic for Occupational, Social and Environmental Medicine, University Erlangen-Nuremberg in Germany. The determination of TCPy, IPPX, 1- and 2-naphthol was carried out using a gas-chromatographic-tandem mass spectrometric (GC–MS/MS) method, which was described in detail elsewhere (Schmidt et al., 2013). In brief, the conjugates of the phenolic structure to glucuronic acid and sulfate were cleaved enzymatically. Afterwards, the phenolic compounds were extracted from the urinary matrix by solid phase extraction and underwent a derivatization with *N*-tert-butyltrimethylsilyl-*N*-methyltrifluoroacetamide (MTBSTFA). Isotope-labeled equivalent structures were used as internal standards for each of the analytes. Urinary concentrations of DMP, DEP, DMTP, DETP, DMDTP, and DEDTP were determined using a GC–MS/MS procedure (Barr et al., 2010; Berman et al., 2013b). Isotope labeled analogues of the analytes were added to the urine, which was then freeze-dried. The lyophilized urine was extracted with diethyl ether and acetonitrile. Then the analytes were derivatized with pentafluorobenzyl bromide. After addition of water, liquid-liquid extraction was carried out twice with hexane to separate the derivatives from matrix components and excess of derivatization agent. Thereafter GC–MS/MS analysis took place. Calibration was performed with standard solutions prepared in pooled urine.

The limit of detection (LOD) and limit of quantification (LOQ) were determined by means of a seven equidistant point calibration near the proposed LOD, according to guideline DIN 32645 (Schmidt et al., 2013). LOD and LOQ were estimated using a signal-to-noise ratio of 3:1 and 9:1, respectively. LOD was 0.2, 0.5, 0.2 and 0.1 µg/L, respectively, and LOQ was 0.7, 1.7, 0.9 and 0.4 µg/L, respectively, for TCPy, IPPX, 1- and 2-naphthol. The LOD ranged from 0.01 µg/L for DEDTP to 0.1 µg/L for DEP, DETP, DMP, DMTP, and DMDTP while the LOQ ranged from 0.03 µg/L or DEDTP to 0.3 µg/L for DEP, DETP, DMP, DMTP, and DMDTP. Creatinine in urine was determined according to the Jaffé method (Larsen, 1972). Quality assessment of the analytical procedures

Download English Version:

<https://daneshyari.com/en/article/6312649>

Download Persian Version:

<https://daneshyari.com/article/6312649>

[Daneshyari.com](https://daneshyari.com)