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Urinary concentrations of environmental contaminants and phytoestrogens in adults in Israel



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ABSTRACT

Background: The Ministry of Health Biomonitoring Study estimated exposure of individuals in the Israeli population to bisphenol A (BPA), organophosphate (OP) pesticides, phthalates, cotinine, polycyclic aromatic hydrocarbons (PAHs), and the phytoestrogenic compounds genistein and daidzein.

Methods: In 2011, 250 individuals (ages 20-74) were recruited from five different regions in Israel. Urine samples were collected and questionnaire data were obtained, including detailed dietary data (food frequency questionnaire and 24 hour recall). Urinary samples were analyzed for BPA, OP metabolites (dialkyl phosphates), phthalate metabolites, cotinine, PAH metabolites, genistein, and daidzein.

Results and discussion: BPA urinary concentrations were above the limit of quantification (LOQ) in 89% of the samples whereas urinary concentrations of phthalate metabolites were above the LOQ in 92–100% of the samples. PAH metabolites were above the LOQ in 63–99% of the samples whereas OP metabolites were above the LOQ in 44–100% of the samples. All non-smoking participants had detectable levels of cotinine in their urine; 63% had levels above the LOQ, and the rate of quantification was high compared to the general non-smoking population in Canada. Median creatinine adjusted concentrations of several OP metabolites (dimethyl phosphate, dimethyl thiophosphate) were high in our study population compared to the general US and Canadian populations. Median creatinine adjusted urinary BPA concentrations in the study population were comparable to those in Belgium and Korea; higher than those reported for the general US, German, and Canadian populations; and very low compared to health-based threshold values. Phthalate concentrations were higher in our study population compared to the general US population but values were very low compared to health-based threshold values. Median creatinine adjusted PAH concentrations were generally comparable to those reported for the general US population; median creatinine adjusted daidzein concentrations were high in our population compared to the general US population whereas genistein concentrations were comparable.

Conclusions: We interpreted observed urinary contaminant levels observed in our study by comparing values with health-based threshold values and/or values from international human biomonitoring studies. Using this data interpretation scheme, we identified two contaminants as being of potential public health concern and high priority for public health policy intervention: environmental tobacco smoke (ETS) and OP pesticides. We used the data collected in this study to support public health policy interventions. We plan to conduct a follow-up biomonitoring study in 2015 to measure ETS and OP exposure in the general population in Israel, to evaluate the effectiveness of relevant policy interventions.

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1. Introduction

Populations worldwide are exposed to a variety of chemical compounds from the environment and consumer products. However, the extent of exposure to these compounds may differ between populations or population groups because of differences in local pollution levels and life-style. Chronic exposure to chemicals in the environment and in consumer products may impact public health, specifically chronic exposure

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to pesticides (McKinlay et al., 2008), phthalates (Meeker et al., 2009), polycyclic hydrocarbons (PAH) (Liu et al., 2008), phenolic compounds like bisphenol A (BPA) (Rubin, 2011) and naturally derived compounds with endocrine activity (phytoestrogens) (Dinsdale and Ward, 2010). Human biomonitoring (HBM) is an essential tool for estimating individual exposure to chemical compounds, so it is used frequently in studies which explore sources and pathways of exposure, and the relationship between disease and exposure to chemicals. As HBM is an important tool for determining the effectiveness of public health efforts to reduce public exposure to specific chemicals, for tracking time trends in exposure levels of the population to environmental chemicals, and for determining whether exposure levels are higher among such potentially vulnerable groups as minorities and children, many countries have developed national biomonitoring programs.

Human biomonitoring studies to date in Israel have focused on "classic" environmental pollutants (heavy metals, organochlorine pesticides, and PCBs) and have been limited by small sample sizes (Berman et al., 2012). The purpose of the Ministry of Health Biomonitoring Study was to estimate exposure to a range of environmental pollutants in the general population in Israel. We measured urinary concentrations of several environmental contaminants (bisphenol A (BPA), organophosphate (OP) pesticides, phthalates, cotinine, polycyclic aromatic hydrocarbons (PAHs)), and the phytoestrogenic compounds genistein and daidzein in a sample of 250 adults from the general population. The objectives of the study included determining the average (and range of) urinary concentrations of environmental chemicals in individuals in the study population and identifying demographic and dietary predictors of exposure to these contaminants in the Israeli population.

The purpose of the paper is to present descriptive data on urinary concentrations of BPA, OP pesticides, phthalates, cotinine, PAHs, and the phytoestrogenic compounds genistein and daidzein in our study population of 250 Israeli adults, and to discuss the environmental health policy implications of the observed chemical concentrations.

2. Materials and methods

2.1. Recruitment

The potential study population included Israeli adults, ages 20–74. Recruitment and interviewing took place between February and June 2011. The parameters for defining the sample were selected so as to represent the population distribution of urban vs rural dwelling (with urban defined as population more than 2000) and the two major ethnic groups in Israel (Jews and Arabs), as well as wide geographical representation. Overall, 20 cities/towns were selected, with 4 representing the Arab sector (3 urban, 1 rural) and the others the Jewish sector (15 urban, 1 rural). In each town/city, interviewers were requested to interview 15 people. Within each city/town, interviewers were required to select 5 separate areas. Within each area, recruitment was done by "knocking on doors" and interviewing those who met the inclusion criteria and agreed to participate, including providing a urine sample. The inclusion criteria were age (20–74) and ability to answer the questionnaire in Hebrew or Arabic. For each address visited, where there were individuals who refused to participate in the study or were not at home, this was documented. The response rate was 29%, excluding individuals not eligible for the study and individuals not home at the time of the visit. Participants were not targeted for specific exposure scenarios (suspected geographic hotspots or occupation).

The study protocol was reviewed and approved by the Sheba Tel Hashomer Helsinki Committee. Written informed consent was obtained from all respondents. Participation in the study was voluntary and study participants received a small gift (coffee mug) in compensation for study participation. At the time of recruitment participants received a letter explaining that they would receive individual results (urinary concentrations of environmental contaminants) if they requested it at the time of their interview or if their individual urinary metabolite

results were unusual (more than 10 times the 90th percentile for the study population). We reported individual results to 18 study participants who requested their individual results and one study participant with unusual urinary BPA concentrations. For individuals who requested results we reported the absolute urinary concentration, the mean urinary concentration in the population, and the range of urinary concentrations in the study population. For the individual with the unusual urinary BPA concentration we included information on possible exposure sources. All individuals receiving results were invited to contact the study coordinator for additional information. All analyses of data for the study were conducted without details on the identity of the participants.

2.2. Questionnaire

A trained interviewer administered a detailed structured questionnaire to the study participants. The questionnaire used in the current study was based on the Israel Health and Nutrition Survey questionnaire. The interviews were carried out in Hebrew or Arabic. The interview included a 24 hour dietary recall, an adapted food frequency questionnaire, a demographic questionnaire and a health/lifestyle questionnaire. The health/lifestyle questionnaire included specific questions on occupation, on consumption of water from water coolers and sports bottles, on frequency of heating food in the microwave, frequency of consumption of grilled foods and smoked foods, use of cosmetics and personal care products, and use of pesticides in the garden, home, and for pets. We adapted the standard validated food frequency questionnaire used in the Israel Health and Nutrition Survey by adding soy products and fruits which we suspected had high levels of organophosphate residues based on pesticide residue surveys conducted by the Ministry of Health.

Socio-demographic and personal variables included age (analyzed as a continuous variable, and in addition grouped as 20–44 years and 45–74 years), gender (males/females), urbanicity (urban/rural), country of birth (Israel/other), education (school level qualification or below [hereafter, lower education]/higher education), and ethnicity (Jewish/Arab and Druze/ Other).

Smoking status was based on self-report. Questions used for active tobacco smoking status were: "Do you currently smoke, including a hookah (water pipe)?", "What do you currently smoke, or what did you smoke before? (cigarettes, cigars, pipe, hookah, other)", and "How many cigarettes do you smoke per day or per week?" Based on the first question, participants were grouped as tobacco smokers (of any kind) or nonsmokers.

2.3. Urine sample collection

Spot urine samples were collected at the time of interview in prescreened BPA and phthalate free 120-mL polypropylene urine containers. All urine samples were maintained at 4 °C for a maximum of 24 h until they were transported to the Sheba Medical Center (SMC) at Tel Hashomer. Urine samples were aliquoted at SMC and frozen at $-20\,^{\circ}\text{C}$. Within four months of collection, urine samples were shipped to the University of Erlangen-Nuremberg in Germany on dry ice $(-70\,^{\circ}\text{C})$, where they were analyzed.

2.4. Sample analysis

Laboratory analyses of environmental chemicals and creatinine were performed at the Institute and Outpatient Clinic for Occupational, Social and Environmental Medicine, University Erlangen-Nuremberg in Germany using the following methods.

For the determination of cotinine in urine a gas chromatographymass spectrometry (GC–MS) procedure was applied (Müller, 2003). For separation of the analyte from the biological matrix the urine was extracted with dichloromethane. Deuterated cotinine was used as an

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