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Environmental and lifestyle factors affecting exposure to polycyclic aromatic hydrocarbons in the general population in a Middle Eastern area

Mohammad Hoseini [a], Ramin Nabizadeh [b, c], Juana Maria Delgado-Saborit [d, e, f, g], Ata Rafiee [c], Kamyar Yaghmaeian [b, c], Saeid Parmy [c], Sasan Faridi [b, c], Mohammad Sadegh Hassanvand [b, c], Masud Yunesian [b, c, **], Kazem Naddafi [b, c, *]

[a] Research Center for Health Sciences, Institute of Health, Department of Environmental Health, School of Health, Shiraz University of Medical Sciences, Shiraz, Iran
[b] Center for Air Pollution Research (CAPR), Institute for Environmental Research (IER), Tehran University of Medical Sciences, Tehran, Iran
[c] Department of Environmental Health Engineering, School of Public Health, Tehran University of Medical Sciences, Tehran, Iran
[d] ISGlobal Barcelona Institute for Global Health, Barcelona Biomedical Research Park, Barcelona, Spain
[e] Pompeu Fabra University, Barcelona, Spain
[f] Spanish Consortium for Research on Epidemiology and Public Health (CIBERESP), Instituto de Salud Carlos III, Madrid, Spain
[g] Division of Environmental Health & Risk Management, School of Geography, Earth & Environmental Sciences, University of Birmingham, United Kingdom

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ABSTRACT

The aim of this study was to investigate environmental and lifestyle factors affecting exposure to PAHs in the general population in a large city of the Middle East (Tehran) by measuring urinary monohydroxy polycyclic aromatic hydrocarbons (OH-PAHs) and establishing relationships between PAHs exposure and related factors. Urine samples were collected from 222 randomly chosen subjects who were living in the urban area of Tehran, Iran. Subjects were required to complete a detailed questionnaire aimed to document their personal and sociodemographic information, activities, cooking-related appliances, smoking history/exposure, and consumed foodstuff. Identification and quantification of six OH-PAHs was carried out using a gas chromatography with mass spectrometry (GC-MS). The geometric means for 1-OHP, 1-NAP, 2-NAP, 2-FLU, 9-FLU, and 9-PHE for whole population study were 310, 1220, 3070, 530, 330, and 130 ng/g creatinine, respectively. The two naphthalene metabolites contributed on average 77% of the total concentration of six measured OH-PAHs, followed by the 2-FLU, 1-OHP, 9-FLU, and 9-PHE. The most important predictors of urinary PAHs were consumption of grilled/barbecued foods, smoking, and exposure to environmental tobacco smoking. Water pipe smoking was linked to urinary OH-PAH metabolite in a dose-response function. Residential traffic was also related with OH-PAH metabolite concentrations. Other factors including gender, age, exposure to common house insecticides, open burning, and candle burning were found to be statistically associated with the urinary levels of some OH-PAHs. High exposure to PAHs among general population in Middle Eastern large cities and its associated health implications calls for public health measures to reduce PAHs exposure.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are widespread environmental carcinogens, mostly formed as a result of incomplete combustion process (Liaud et al., 2015; Wang et al., 2014). Sources of PAHs can be both natural and anthropogenic, with the most common sources being industrial plants, motor vehicle exhaust, residential heating with fossil fuels, wood-burning ovens and fireplaces, cigarette smoke, forest and grass fires, and charcoal-

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* Corresponding author. School of Public Health, Poursina Avenue, Qods Street, Enqelab Square, Tehran, Iran.
** Corresponding author. School of Public Health, Poursina Avenue, Qods Street, Enqelab Square, Tehran, Iran.
E-mail addresses: yunesian@tums.ac.ir (M. Yunesian), knadafi@tums.ac.ir (K. Naddafi).

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grilled and flame-broiled food. Even more, a new trend of exposure to PAHs is recently emerging and spreading fast worldwide linked with smoking water pipes (Gurung et al., 2016). This exposure is becoming common especially among the young population (Gurung et al., 2017). Therefore PAH exposure in community settings occur on a regular basis for most people via different sources, including air, food, soil, and water (Abdel-Shafy and Mansour, 2016; Hoseini et al., 2016a). Several of the PAHs, including benzo[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, chrysene, dibenz[a,h]anthracene, and indeno[1,2,3-c,d]pyrene, have been shown to be human carcinogens and can cause several adverse effects including lung, gastric, skin, prostate, and other cancers (Rengarajan et al., 2015).

Due to carcinogenic, mutagenic, and teratogenic properties of PAHs, there is an growing interest in evaluating the body burdens of PAHs absorbed by different exposure routes, including inhalation (Li et al., 2008), ingestion (Rey-Salgueiro et al., 2009a, 2009b), and dermal contact (Okuda et al., 2010). To achieve this goal and obtain estimates of the total PAH exposure and the internal dose, PAHs and their metabolites should be quantified in the body fluids. Once in the body, PAHs are bio-transformed and metabolized in two phases. Metabolic activation of PAHs starts with oxidizing PAHs via the cytochrome P450 enzymes (mainly CYP1A1) to produce more reactive metabolites, which are further changed to diol-epoxides that can initiate cell transformation. In the next phase, the products formed previously are combined with endogenous hydrophilic compounds such as glucuronic acid and sulfate to create compounds with sufficient hydrophilic character to allow excretion through urine or feces (Estabrook et al., 1981; Li et al., 2008; Xue and Warshawsky, 2005).

Urinary hydroxy-PAHs (OH-PAHs) have been proposed as biological markers of individual internal dose to assess total PAH exposure. Among these hydroxy-PAH metabolites, 1-hydroxypyrene (1-OHP), the metabolite of pyrene, has been used as the most common indicator of exposure to total PAHs in biomonitoring programs. However, based on the fact that PAHs are emitted through different sources and absorbed via various routes, pyrene could not be an appropriate marker for estimating total PAHs intake. So, the measurement of other PAHs metabolites along with 1-OHP can be a useful way to provide information on recent exposure to total PAHs (Gunier et al., 2006; Jongeneelen, 1994; Li et al., 2008, 2016).

Many studies have been conducted to assess occupational PAH exposure by measuring levels of urinary OH-PAHs in various occupational settings. Moreover, there are several investigations which have been conducted to address the urinary levels of OH-PAHs in various specific sub-populations (Adetona et al., 2013; Wei et al., 2016), and people living near some industries and certain facilities (Lu et al., 2016; Ranzi et al., 2013; Srogi, 2007). Nowadays, however, researchers are mainly focusing on low-level, environmental exposure to PAHs in the general population. Biomonitoring of PAHs in the general population has been conducted in few countries such as the United States (Grainger et al., 2006; Li et al., 2008), Germany (Scherer et al., 2000), Australia (Thai et al., 2016), Canada (Viau et al., 1995), Ukraine (Mucha et al., 2006), United Kingdom (Aquilina et al., 2010) and Korea (Sul et al., 2012). There are, however, lack of any study regarding assessment of PAH exposure in general population in a Middle Eastern country, such as Iran, with meteorological conditions favouring gas phase PAHs and subsequent inhalation intake; widespread presence of grilled and charcoal food (e.g. kebabs) favouring dietary intake of PAHs; and widespread traditional socializing activities such as water-pipe smoking, which enhance the exposure to PAHs to the general population. Considering this fact and also the hazards associated with exposure to PAHs, the present study was conducted to investigate the levels and concentration profiles of different OH-PAHs excreted in urine in a Middle Eastern country. In addition, the influences of different predicating factors such as age, gender, diet, smoking patterns, including water-pipe smoke, and other routine activities on the levels of these PAH metabolites were studied. Indeed, this is the first study evaluating any relationship between water-pipe smoke and presence of urinary PAH biomarkers as a tracer to PAH exposure.

2. Materials and methods

2.1. Study subjects and sample collecting protocol

This study was conducted among the general population of Tehran, which is the capital of Iran and one of the most populated cities in the Middle East and also in the world. The city has almost 12 million inhabitants (as per 2014) and is situated in the northern part of Iran (Naddafi et al., 2011; Kamani et al., 2014). The study population consisted of 222 randomly chosen subjects which were recruited by conducting door knock surveys. It should be noted that from the timing of sampling, the first urine samples, which could be the most concentrated were not included anyway. Samples were normalized with the creatinine concentration to account for any dilution effect. The sampling procedure was started with the selection of random points as described in our previous study (Hoseini et al., 2016b). In brief, after gathering, samples were transferred to the laboratory in dry ice, at about 2–8 °C. All subjects were requested to fill a detailed questionnaire (See Supporting Information) which included different parts such as demographic information, activities, occupational exposure, smoking habits, food cooking appliances, residential environment characteristics, and eating habits during the week prior to sampling. Ethical approval for this study was issued by the Ethics Committee of Tehran University of Medical Sciences (code No 24039).

2.2. Laboratory analysis of PAH metabolites

The measurement of six OH-PAH metabolites including 1-Hydroxy pyrene (1-OHP), 1-Hydroxy naphthalene (1-NAP), 2-Hydroxy naphthalene (2-NAP), 2-Hydroxyfluorene (2-FLUO), 9-Hydroxyfluorene (2-FLUO), and 9-Hydroxyphenanthrene (9-PHEN) were carried out by enzymatic hydrolysis and solid phase extraction (SPE). Quantification of metabolites was performed by gas chromatography/mass spectrometry (GC/MS) according to the procedure developed by Romanoff et al. (2006) with some modification. In brief, urine samples (3 mL) were transferred to test tubes (16 mm × 100 mm) and spiked with 10 μL of the internal standard solution (naphthalene-d8 and phenanthrene-d10). Next, sodium acetate buffer (0.1 M; pH 5.5; 5 mL) and β-glucuronidase/arylsulfatase enzyme (10 μL, Helix pomatia, H-1; Roche Diagnostic) were added to each sample. After that, the samples were tighten with Teflon-lined screw caps, mixed and then incubated at 37 °C for 17–18 h (overnight) at 210 rpm. After enzymatic hydrolysis, the samples were mixed, allowed to equilibrate and then applied to a SPE column packed with 0.5 g RP-C18 (Applied Separation, USA) using an Extraction Manifold (20 positions, 13 × 75 mm tubes, Waters) connected to a vacuum pump. Cartridges were conditioned with 3 mL methanol and 3 mL purified water, and then samples were injected (1 mL/min) to the column. After washing with purified water (3 mL, 10 mL/min) and methanol/sodium acetate buffer (3 mL, 4:6 v/v, pH 5.5, 10 mL/min), the cartridges were dried under a gentle flow of nitrogen for 10 min and the final extract was eluted with 3 mL of dichloromethane (0.5 mL/min). Then, 5 μL of dodecane was added to extracts and evaporated to about 5 μL under a stream of nitrogen at 40 °C and the residues were dissolved in 100 μL