



Environmental determinants of polycyclic aromatic hydrocarbons exposure at home, at kindergartens and during a commute



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ABSTRACT

The aim of this study was to assess the potential health risk of exposure to polycyclic aromatic hydrocarbons (PAHs) at home and kindergarten for pre-school children. The urine samples were taken from 200 pre-school children aged 5–7 years and analyzed for 1-OHP as a biomarker of PAHs. Mixed effect models were applied to investigate the association between effective environmental parameters (mode of transport, distance to major roads, traffic density, greenness, tobacco exposure, home ventilation, and grill foods) and urinary 1-OHP levels. A Monte-Carlo simulation technique was applied to calculate the risk of exposure to PAHs and to check the uncertainty of input variables and the sensitivity of the estimated risk. The median and inter quartile range (IQR) of 1-OHP was 257 (188.5) ng L⁻¹. There was a positive significant association between distance from the kindergartens to the green space with surface area ≥ 5000 m² and 1-OHP concentration ($\beta = 0.844$, 95% CI: 0.223, 1.46, P-value = 0.009). Also, urinary 1-OHP was found to be inversely associated with the time the window was open at the home ($\beta = -12.56$, 95% CI: -23.52, -1.596, P-value = 0.025) and normalized difference vegetation index (NDVI) in a 100 m buffer around the homes. The mean (9.76 E-3) and 95th percentile (3.28 E-2) of the hazard quotient (HQ) indicated that the concentration of urinary 1-OHP is at a safe level for the target population (HQ < 1). According to the sensitivity analysis results, the concentration of 1-OHP is the most influential variable in the estimated risk. Our findings indicated that the proximity of homes and kindergartens to green space areas and their remoteness from the main streets and heavy traffic areas are associated with reduced exposure to PAHs.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are products of incomplete combustion of fuels in the industrial and non-industrial activities (Mucha et al., 2006). The most common sources of exposure to PAHs are vehicle exhausts, cigarette smoking, wood burning, coal firing, steel plants, and grilled and smoked foods (Kim et al., 2013). In addition to inhalation route of exposure, PAHs are also found in soil and water resources. Due to more activity and consequently higher inhalation rate, children are more exposed to PAHs than adults (Kirby, 2003; Landrigan et al., 1998). In a similar condition, PAHs metabolites

in children are approximately 1.3 times that of adults (Fan et al., 2012). Pyrene is the most common type of PAHs and its urinary metabolite, 1-hydroxypyrene (1-OHP), is usually used as a biomarker of exposure to total PAHs (Ruíz-Vera et al., 2015; Strickland and Kang, 1999). The 1-OHP is accepted as a valid biomarker for short-term exposures to PAHs (Ciarrocca et al., 2014). When pyrene enters the body, it is first metabolized by cytochrome P450 1A1 (CYP1A1) enzymes and it then appears as 1-OHP in the urine (Jongeneelen, 2001; Zare et al., 2013). The half-life of 1-OHP has been reported to be between 6 and 35 h (Brzeźnicki et al., 1997; Buchet et al., 1992; Jongeneelen et al., 1990).

Most studies on exposure to PAHs in children are about dietary

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intake, environmental tobacco smoke (ETS), and traffic related air pollutants (TRAP). However, TRAP is reported as the most important source of 1-OHP in urine samples of children (Conte et al., 2016; Kanoh et al., 1993; Morgan et al., 2015; Perez-Maldonado et al., 2017; Shahsavani et al., 2017; Thai et al., 2016; Yu et al., 2016). Risk assessment using deterministic models, based on average values or extreme cases, leads to under or overestimated risk values (Lonati and Zanoni, 2012). In addition, use of deterministic models does not include the role of parameters variability in the calculated risk values. Use of probabilistic risk assessment methods by stochastic modeling of exposures is an alternative to classic deterministic approach. Monte-Carlo simulation technique is the most common probabilistic approach for health risk assessment, due to its accuracy and reliability (Lonati et al., 2007). This approach use parameters distribution instead of fixed values of the parameter, and finally calculates the probability distribution function of the output. This approach has been successfully applied in several studies for assessment of cancer and non-cancer risk of exposure to chemicals in water, air soil and food (Fallahzadeh et al., 2018; Miri et al., 2016b, 2017a; Ruiz-Vera et al., 2015).

The main objective of this study was to quantify the exposure of preschool children to PAHs by measuring urinary 1-OHP levels in urine samples. The impacts of different environmental determinants such as exposure to tobacco smoke, traffic related pollutants TRAP, and the greenness on the urinary 1-OHP of children were also investigated. Probabilistic health risk assessment based on Monte-Carlo simulation technique was also applied to quantify the health risk imposed by PAHs exposure in children.

2. Material and methods

2.1. Study area

The study was conducted in the Sabzevar city, northeastern part of I.R. Iran in Khorasan Razavi province. The population of the city was about 240,000 according to 2016 census. The area of the city is about 23 km². It's located in the hot and dry region (coordinates: 36°12'N 57°35', elevation: 977.6 m) with arid climate. The annual average rainfall in Sabzevar is 330 mm, its average moisture content is 43%, and wind usually blows from the east to west of the city. The annual minimum, maximum, and average temperature in the city are −22, 45, and 16 °C, respectively. Sabzevar is located beside the most congested highway in Iran (Tehran to Mashhad) and suffering from air pollution problems. Fig. 1 shows the study area and selected kindergartens.

2.2. Study design and sampling

As a first step, 60 kindergartens were randomly selected throughout the city. All children aged between 5 and 7 years were invited to participate in the study (about 900 children). Finally, the parents of 200 children from 27 kindergartens agreed to contribute their children to the program. The average number of participants in each of the selected kindergartens was 7. All of them were in the kindergarten at least 1 year before the study and their homes have not changed since their birth. The study design was approved by the Clinical Research Ethical Committee (IR.SSU.SPH.REC.1395.66) of the Shahid Sadoughi University of Medical Science, Yazd, Iran. All parents signed an informed consent form.

The urine samples were taken in June 2017. About 20 mL urine sample was collected in the morning. Urine samples were transferred to the laboratory within less than 2 h. Samples were transferred into 50 mL conical polypropylene tubes covered with aluminum foil and stored in −80°C until analysis.

A questionnaire was designed to obtain the required information such as age (year), gender (male/female), weight (kg), body mass index (BMI), ventilation at home (hood application Yes/No), cooking time (h day^{−1}) at home, window opening time for natural ventilation

(h day^{−1}), exposure to tobacco at home (Yes/No), the number of cigarette smoked at home per day, out-door exposure to tobacco (Yes/No), out-door exposure duration to tobacco (h/week), commuting to kindergarten (walking, motorcycle, car, and bus), and eating grilled foods in the day before sampling (Yes/No). The distance between school or home to the major roads and the total length of the streets in the 100 m buffer around the homes and kindergartens have been used as a surrogate of traffic intensity (Amini et al., 2016; Henderson and Brauer, 2005). The average of the normalized difference vegetation index (NDVI) values in the 100 m buffer around the children's home and kindergartens were used as a measure of greenness (Dadvand et al., 2012a, 2015a). NDVI was calculated based on images obtained from Landsat 8 satellite at 30 × 30 m resolution (Triguero-Mas et al., 2015). The Landsat 8 satellite consists of two science instruments called Operational Land Imager (OLI) and the Thermal Infrared Sensor (TIRS). These sensors provide seasonal coverage of the global landmass at a spatial resolution of 30 m (visible, NIR, SWIR), 100 m (thermal), and 15 m (panchromatic). NDVI map was produced using the images obtained on June 15, 2017. Before NDVI calculation, radiometric corrections were made on the image. The distance between homes and kindergartens to any green space and the green spaces with a surface area over 5000 m² were also calculated (Dadvand et al., 2012a, 2016). NDVI is an indicator of green vegetation density and it is calculated based on the land surface reflectance of visible red and near-infrared parts of the spectrum (Dadvand et al., 2015b). The ranges of NDVI values are between −1 and 1. A higher NDVI values indicate higher greenness.

2.3. Measurement of 1-OHP

Urinary 1-hydroxypyrene concentration was measured according to Jongeneelen et al. (1987) and Brucker et al. (2013) method. In brief, 5 ml of 0.1 mol L^{−1} acetate buffer (pH 5) was added to 2.5 ml of urine sample to make a final volume of 7.5 ml. Then, 10 μL of glucuronidase-arylsulphatase was added to this solution and incubated for 2 h at 37 °C in a shaking bath. PAHs metabolites were extracted from the urine samples using a solid phase extraction (SPE) method on a C18 cartridge (CHROMABAND® C18ec 3 ml, 500 mg, 50/pk). Two milliliters of methanol followed by 5 mL of distilled water was applied to activate the cartridges. After sorbent activation, the urine samples were loaded on cartridges at a flow rate of 10 ml min^{−1}. Then, 6 ml of 40% methanol was passed through the cartridges to wash it. Finally, 1-hydroxypyrene was eluted using 2 ml isopropanol. Before injection to high performance liquid chromatography (HPLC), final solvent was evaporated at 37 °C and reconstituted by 200 μl methanol.

The concentration of 1-hydroxypyrene was determined using KNAVER (Germany) HPLC equipped with a C18 reversed phase column (250 × 4.6 mm, with 5 μm particle size; Germany). A mixture of 40% methanol and 60% acetonitrile (40:60 v/v) at a flow rate of 1 ml min^{−1} was used as mobile phase. The fluorescence detector was programmed to operate at an excitation wavelength of 242 nm and an emission wavelength of 388 nm. The limit of detection (LOD) and the limit of quantification (LOQ) of the method was 3 and 10 ng L^{−1} respectively. The standard curve constructed by 8 serial concentrations of 1-OHP (from 10 to 10 × 10⁶ ng L^{−1}) dissolving 1-OHP (Sigma Aldrich, Germany) standard in dichloromethane. All samples were injected in duplicate. The 1-OHP concentration in all samples was above than the LOD.

2.4. Risk assessment

Non-carcinogenic risk of exposure to pyrene was calculated as an important part of PAHs (Yuan et al., 2015). Daily exposure dose of pyrene (EDI in ng/kg-day) was estimated using the following equation (Lakind and Naiman, 2008; Ruiz-Vera et al., 2015).

$$EDI = (C \times P \times EF \times ED) / (BW \times AT \times F) \quad (1)$$

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