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Levels of 1-hydroxypyrene and other monohydroxy polycyclic aromatic hydrocarbons in children: A study based on U.S. reference range values

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Abstract

Urine samples collected in 1999 and 2000 as part of the National Health and Nutrition Examination Survey (NHANES) were analyzed for 14 monohydroxy polycyclic aromatic hydrocarbons (PAH, metabolites of 7 PAH compounds) and for the first time reference range values were calculated for these metabolites in the U.S. population. The purpose of this paper is to explore differences in these PAH metabolites between children (6–11 years old), adolescents, and adults. More than 99% of the urine samples contained a detectable amount of 1-hydroxypyrene (1-OHpyrene), a metabolite of pyrene. We found that children in the youngest age group (6–11 years) had a geometric mean level (creatinine corrected data) 30% higher than children and adults in the other age groups, but no statistical differences existed between the two genders and among different racial groups. Smokers and persons exposed to environmental tobacco smoke (ETS) in 12–19-year-old group and the 20-year-and-older group had higher levels of urinary 1-OHpyrene by a factor of 2–3 than non-smokers in the corresponding age group. Measurements of 3-hydroxyphenanthrene also suggested increased levels in children and in smokers. These results may indicate that young children are at a greater risk for PAH exposure, or that they absorb, distribute, metabolize, or eliminate PAH differently than adults.

Keywords: Pyrene; PAH exposure; Reference range; Children; Urine; GC/MS

1. Introduction

Pyrene has been studied extensively in the last two decades, and is a product of incomplete combustion; its major sources include forest fires, diesel exhaust, road pavement, roofing materials, charcoal-grilled food, and cigar and cigarette smoking (IARC, 1973, 1984, 1985, 1987). Human exposure usually is assessed by monitoring 1-hydroxypyrene (1-OHpyrene) in urine samples because pyrene is the major component in PAH mixtures and its urinary metabolites (1-OHpyrene and 1-OHpyrene glucuronide) usually are abundant and relatively easy to measure. Previous studies found elevated 1-OHpyrene levels in smokers (Gundel et al., 1996; Li et al., 2000), people living in certain industrial regions (Gundel et al., 1996; Zhao et al., 1990), and workers exposed to coal-tar products (Jongeneelen et al., 1985),

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but little is known about the range of levels among non-occupationally exposed populations.

Smoking habits are the most important factor influencing urinary 1-OHpyrene levels in the general population. More than a decade ago, Jongeneelen et al. (1990) monitored a group of coke-oven workers and found that within the control group the smokers' mean urinary 1-OHpyrene level was two times higher than that of the non-smokers. A number of independent studies have confirmed this result in the last decade (Viau et al., 1995; Gundel et al., 1996; Goen et al., 1995; Li et al., 2000). In a study of 124 housewives in Bottrop, Germany, Gundel et al. (1996) pointed out that the influence of smoking was of such a magnitude that potential environmental exposure to PAH in this highly industrialized area could have been obscured by smoking habits. Li et al. (2000) also found that the urinary 1-OHpyrene concentration correlated strongly with the number of cigarettes smoked; in some heavy smokers' urine, 1-OHpyrene concentrations could be 30 times higher than in urine of non-smokers. A few other studies that focused on occupational exposure also reported higher levels of urinary 1-OHpyrene in smokers than in non-smokers when exposed to PAH in the same working environment (Sherson et al., 1992; Granella and Clonfero, 1993; Levin et al., 1995; Vanschooten et al., 1995; Gilbert and Viau, 1997; Dor et al., 2000).

Studies focusing on children's exposure to PAH were published about a decade ago, and levels of urinary 1-OHpyrene varied widely. The low end was around 0.01 μmol/mol creatinine, close to the instrument detection limit (Chuang et al., 1999; Fiala et al., 2001), while the high end was a few hundred times greater, 3–7 μmol/mol creatinine (van Wijnen et al., 1996; Siwinska et al., 1998).

Several studies have assessed uptake pathways. The studies of van Wijnen et al. (1996) and Fiala et al. (2001) analyzed soil, air, food and beverages and concluded that daily diet dominated other environmental sources for children's PAH uptake. Wilson et al. (2000) suggested that PAH uptake occurred mostly through inhalation, followed by dietary ingestion and then non-dietary ingestion. In addition to the normal sources for PAH in adults, children were also exposed to these compounds from soil in the playground.

A few studies have shown that children from low-income families have greater exposure to PAH because they were more likely to be exposed to air polluted by heavy traffic, industrial sources, cigarette smoking, coal-burning stoves, and contaminated house dust or soil. Kanoh et al. (1993) and Fiala et al. (2001) demonstrated that children in the highly polluted areas had urinary 1-OHpyrene levels significantly higher than children in less-polluted areas. Siwinska et al. (1999) found that the urinary level of 1-OHpyrene tended to increase in children exposed to environmental tobacco smoke (ETS). The same group also reported daily median urinary 1-OHpyrene concentration in children of smoking mothers up to 80% higher than it was in children of non-smoking mothers (Siwinska et al., 1998). Wilson et al. (1999) found that the mean concentration of PAH in the play area soil at day care centers serving primarily low-income clients was as much as 42 times higher than at higher-income centers. Furthermore, Fiala et al. (2001) found significantly higher concentrations of urinary 1-OHpyrene in children from kindergartens where air and soil PAH concentrations were 3–6 times higher than similar but less polluted kindergartens.

Investigations have not shown significant differences between children and adults exposed to the same living environment. In a study of PAH exposure of children in low-income families, Chuang et al. (1999) found no difference in urinary 1-OHpyrene concentrations between children and adults. A recent study by Heudorf and Angerer (2001) included 1213 people (261 children ages of 6–12 years) living in 511 households in Frankfurt, Germany. The urinary 1-OHpyrene levels in these children did not differ significantly from levels in the other age groups.

The goal of the present study was to measure 1-OHpyrene in urine samples collected from a representative sampling of the U.S. population to determine a reference range. We first developed a sensitive method to measure urinary monohydroxy-PAH (OHPAH) at low ppt levels (Smith et al., 2002a), then established an improved protocol for quantitative analysis and quality assurance of OHPAH measurements (Huang et al., 2002a). We then analyzed 2312 urine samples and used the results to establish the first U.S. reference range of OHPAH as biomarkers to assess human exposure to PAH.

2. Materials and methods

2.1. Sample collection

Samples were collected as part of the NHANES 1999 and 2000 (CDC/DHHS, 2003a, 2003b), which was designed in part to assess the U.S. population's exposure to environmental chemicals by using biomonitoring. Beginning in 1999, NHANES became a continuous and annual survey. The sampling plan follows a complex, stratified, multistage, probability-cluster design to select a representative sample of the civilian, non-institutionalized U.S. population. The NHANES protocol includes a home interview followed by