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Korea National Survey for Environmental Pollutants in the human body 2008: 1-hydroxypyrene, 2-naphthol, and cotinine in urine of the Korean population

Donggeun Sul [a], Ryoungme Ahn [b], Hosub Im [c], Eunha Oh [d], Jae Hyoun Kim [b], Jin Gyoung Kim [e], PanGyi Kim [f], Hyoung-Ah Kim [g], Won Yeul Park [h], Bu Soon Son [i], Daeyewn Shin [j], Ae-Sug Shim [k], Wonho Yang [l], Seung-Do Yu [m], Kyoung-Ho Lee [n], Kee Jae Lee [o], Sun-dong Lee [p], Jong Wha Lee [i], Chae Kwan Lee [q], Bong-Ki Jang [i], Kyungho Choi [r], Don-Hee Han [s], Moon-Young Hwang [m], Jin Heon Lee [t,*]

[a] Graduate School of Medicine, Korea University, Seoul, Republic of Korea
[b] Department of Health Science, School of Natural Science, Dongduk Women's University, Seoul, Republic of Korea
[c] Research Institute of Health Sciences, Korea University, Seoul, Republic of Korea
[d] Center for Life and Environmental Science, Neodin Medical Institute, Seoul, Republic of Korea
[e] Department of Business Administration, College of Humanities and Social Sciences, Kongju National University, Gongju, Republic of Korea
[f] Deparment of Occupational and Environmental Health, College of Natural Science, Yongin University, Yongin, Republic of Korea
[g] College of Medicine, The Catholic University of Korea, Seoul, Republic of Korea
[h] Research Department, Hyundae Research Institute, Seoul, Republic of Korea
[i] Department of Environmental Health Sciences, College of Natural Science, Soonchunhyang University, Asan, Republic of Korea
[j] Deparment of Environmental Engineering, College of Engineering, Chosun University, Gwangju, Republic of Korea
[k] Research and Clinical Trial Laboratory Service, Neodin Medical Institute, Seoul, Republic of Korea
[l] Department of Occupational Health, College of Natural Science, Catholic University of Daegu, Daegu, Republic of Korea
[m] Environmental Health Research Division, National Institute of Environmental Research, Ministry of Environment, Incheon, Republic of Korea
[n] Health Group Environmental Safety Team, Semiconductor Samsung Electronics, Yongin, Republic of Korea
[o] Department of Information and Statistics, College of Natural Science, Korea National Open University, Seoul, Republic of Korea
[p] School of Oriental Medicine, Sangji University, Wonju, Republic of Korea
[q] Department of Occupational and Environmental Medicine, College of Medicine and Institute, Inje University, Pusan, Republic of Korea
[r] Department of Environmental Health, Graduate School of Public Health, Seoul National University, Seoul, Republic of Korea
[s] Department of Occupational Health and Safety Engineering, College of Health Sciences, Inje University, Gimhae, Republic of Korea
[t] Department of Environmental Education, College of Education, Kongju National University, 182 Shingwan-Dong, Gongju-Si, Chungnam 314-701, Republic of Korea

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ABSTRACT

The Korea National Survey for Environmental Pollutants in the human body conducts representative Korean population studies, which were first initiated in 2005 in Korea. This study was conducted from 2008 to 2009 to determine the exposure levels of polycyclic aromatic hydrocarbons and nicotine in the Korean general population. The study population consisted of 4702 adult subjects from 196 sampling locations including coastal, rural, and urban areas. The urinary levels of 1-hydroxypyrene, 2-naphthol, and cotinine were measured for exposure of polycyclic aromatic hydrocarbons and nicotine. The geometric means of the urinary 1-hydroxypyrene, 2-naphthol and cotinine concentrations in the Korean general population were 0.15 μg/L (95% confidence interval (CI): 0.13–0.17), 3.84 μg/L (95% CI: 3.57–4.11) and 47.42 μg/L (95% CI: 40.52–54.32) respectively. When these values were compared with reference ranges for the United States and Germany, the levels of 1-hydroxypyrene, 2-naphthol, and cotinine were very similar for Korea and Germany, however, these levels were slightly lower in the United States. This study is the first nationwide survey of exposure to polycyclic aromatic hydrocarbons and nicotine in Korea and provides a background reference range for exposure to polycyclic aromatic hydrocarbons and nicotine in the Korean general population.

1. Introduction

Humans are exposed to a wide range of environmental pollutants and environmental health sciences focus on the link between the presence of contaminants in the environment, and their relationship

* Corresponding author. Fax: +82 41 850 8810.
E-mail address: ejhl@kongju.ac.kr (J.H. Lee).

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to possible adverse health effects (Smolders et al., 2009). Over the past 10 years, advances in analytical sensitivity and an increasing social awareness towards pollutants in the environment has led to more widespread use of biomonitoring as a tool for studies of general population groups (Hays and Aylward, 2009). Human biomonitoring especially applies to metals, polycyclic aromatic hydrocarbons (PAHs), phthalates, dioxins, and pesticides, as well as for aromatic amines, perfluorinated chemicals, environmental tobacco smoke, and volatile organic compounds. Blood and urine are by far the most approved matrices (Angerer et al., 2007).

PAHs are a class of stable organic molecules composed of carbon and hydrogen. PAHs are ubiquitous environmental pollutants in the air, water, and soil, which are produced as a consequence of the incomplete combustion of organic materials. Residential heating sources, refuse burning, and vehicle exhausts also contribute to the widespread distribution of PAHs (Jongeneelen, 2001). The International Agency for Research on Cancer has characterized PAHs as carcinogens (IARC, 1998). Pyrene is a common compound in various PAH mixtures that is metabolized to the intermediary 1-hydroxypyrene (1-OHP), which is further metabolized to 1-OHP-glucuronide (Moen et al., 1996). A recent comprehensive review showed that urinary 1-OHP could be used as a biomarker of environmental and occupational exposure to PAHs (Hansen et al., 2008). Naphthalene is another major compound of PAHs that is metabolized to over 30 different metabolites. Among those, 1- and 2-naphthol have been used as urinary biomarkers of exposure to PAHs (Preuss et al., 2004).

In general, tobacco addiction is considered a public health priority. Moreover, cigarette smoking currently affects 1.3 billon people worldwide and is responsible for approximately 5 million deaths per year (Esson and Leeder, 2004). Indeed, tobacco use is the most important preventable cause of premature morbidity and mortality in Korea and smoking has been found to increase the risk for cancer, emphysema, cardiovascular disease, and possibly other disorders (Arabi, 2006; Eliasson, 2003; Mazza et al., 2010). Cotinine is a metabolite of nicotine that is currently regarded as the best biomarker of nicotine in active smokers and in nonsmokers exposed to environmental tobacco smoke. Measuring cotinine is preferred over measuring nicotine, because cotinine persists longer in the body (Benowitz, 1996).

Recently, the Korea National Survey for Environmental Pollutants in the Human Body has been conducted to determine human exposure to specific environmental pollutants in the general population. The present report describes urinary levels of 1-OHP and 2-naphthol, as exposure biomarkers of PAHs cotinine as an exposure biomarker of nicotine, in the Korean general population.

2. Materials and methods

2.1. Study design

The Korea National Survey for Environmental Pollutants in the Human Body was the preliminary program used to investigate how nationwide human biomonitoring will progress in Korea. This was designed with reference to the National Human Exposure Assessment Study (Sexton et al., 1995) and the German Environmental Survey (Seifert et al., 2000) and was the pilot program for preparing the frame and contents of the basic survey of national environmental health in Korea. The sample units as clusters, which were small geographic units that contained approximately 60 dwellings. The Korea National Survey for Environmental Pollutants in the Human Body population consisted of 196 general sample units, including coastal, rural, and urban areas. A total of 4702 adult subjects whose ages ranged from 20 to 79 years were recruited. All subjects were asked to complete a questionnaire, which was composed of seven parts (including lifestyle, occupational exposure, usual dietary habits, women's history, food intake during the last year, demographic characteristics, and residential environment) and 148 questions (including main and detailed questions). During progression of the Korea National Survey, we attempted a variety of methods to increase the response rate in our programs. The systems were improved to reduce respondent burden, such as distance, time and questionnaire, and the home visiting teams operated to collect human specimens at home anytime, if required by the subjects. In addition, the interviewers were well educated, had a variety of training courses, and were friendly, persuasive, and persistent (but non-confrontational) with respondents. They presented the study in a positive manner that encouraged respondents to cooperate, and informed the subjects about the objectives and sponsors of the study as well as, about the planned uses of the data. They were also sensitive to respondents' concerns and responded accurately, honestly, and reassuringly to respondents' questions and concerns about matters such as respondent privacy, respondent burden, and any risks associated with participation. This study was approved by the institutional review board at the National Institute of Environmental Research and each subject gave written informed consent.

2.2. Sample collection

All urine samples (45 mL) were collected in 50 mL conical tubes that were wrapped in aluminum foil to avoid light exposure and labeled with the subject identification number and date. Following collection, the samples were kept in a portable and insulated bag with ice packs (at about 2–8 °C) and delivered to an analytical laboratory. The sample delivery was conducted using hand-to-hand and cold chain systems.

2.3. Analysis of 1-OHP

1-OHP in urine was analyzed according to the method developed by Jongeneelen et al. (1985) and Kim et al. (1999). All analytical procedures were conducted in a dark room. Briefly, urine samples were heated at 37 °C until completely thawed. Urine samples were spiked with 0.5 mL of 1-OHP stock solution or left unspiked, after which they were shaken and transferred to a 4 mL amber glass vial. Next, 100 μL of 2 M acetate/acetic acid buffer (pH 5) and 10 μL of β-glucuronidase/arylsulphatase (type H-2 from Helix pomatia: activity 110,000 and 1000 U/mL, Sigma Co. St. Louis, MO, USA) were added. The samples were then sealed with Teflon-lined screw caps, gently mixed and then incubated at 37 °C for 16 h in a dry bath while shaking at 180 rpm. After enzymatic hydrolysis, 150 μL of acetonitrile was added, and the samples were centrifuged at 3000 rpm for 5 min. Next, 100 μL volume of the supernatant was injected into a High Performance Liquid Chromatography–fluorescence detector system equipped with a C18 column (ZORBAX SB C18, 5 μm, 4.6 mm inner diameter and 250 mm long, Agilent, USA) that was held at 40 °C. The mobile phase was a mixture of Milli-Q water and acetonitrile (60:40, v/v), which was applied at a flow-rate of 1 mL/min. The analyte was detected using a fluorescence detector at an excitation wavelength 242 nm and an emission wavelength 388 nm. The retention time for 1-OHP was 10.5 min and the detection limit of 1-OHP in urine should have been 0.01 μg/L (signal-to-noise=3). The validation data, which consisted of relative standard deviations, were in the range of 0.3% to 5.3% and 0.5% and 9.9% for within-day and between-day imprecision, respectively.

2.4. Analysis of 2-naphthol

Urinary 2-naphthol was also analyzed according to the method developed by Jongeneelen et al. (1985) and Kim et al. (1999). All analytical procedures were conducted in a dark room. Briefly, urine samples were heated at 37 °C using a water bath until completely thawed. The samples were then spiked with 0.5 mL of 2-naphthol stock solution or left unspiked, after which they were shaken and transferred to a 4 mL amber glass vial, Next, 100 μL of 2 M acetate/acetic acid buffer (pH 5) and 10 μL of β-glucuronidase/ arylsulphatase (type H-2 from Helix pomatia: activity 110,000 and 1000 U/mL, Sigma Co. St. Louis, MO, USA) were added. The samples were then sealed with Teflon-lined screw caps and subsequently gently mixed and incubated at 37 °C for 16 h in a dry bath while shaking at 180 rpm. Following enzymatic hydrolysis, 1.5 mL of acetonitrile was added and the samples were centrifuged at 3000 rpm for 5 min. Next, 20 μL of supernatant was injected into a High Performance Liquid Chromatography–fluorescence detector system equipped with a C18 column (ZORBAX SB C18, 5 μm, 4.6 mm inner diameter and 250 mm long, Agilent, USA) that was held at 40 °C. The mobile phase was a mixture of Milli-Q water and acetonitrile (32:68, v/v), which was applied at a flow-rate of 1 mL/min. The analyte was detected using a fluorescence detector at an excitation wavelength 227 nm and an emission wavelength 355 nm. The retention time for 2-naphthol was 29.4 min and the detection limit of 2-naphthol in urine should have been 0.07 μg/L (signal-to-noise=3). The validation data, which consisted of the relative standard deviations, were in the range of 0.2% to 2.1% and 0.4% and 13.3% for within-day and between-day imprecision, respectively.