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Source attribution of personal exposure to airborne polycyclic aromatic hydrocarbon mixture using concurrent personal, indoor, and outdoor measurements

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ABSTRACT

Objectives: Relative importance of multiple indoor and outdoor venues on personal exposure concentrations to pro-carcinogenic polycyclic aromatic hydrocarbons (c-PAHs) remains poorly understood. This is particularly challenging because many c-PAHs share sources and occur as a complex mixture. Accurate and precise apportionment of personal exposure according to exposure venues could aid in the understanding of human health effects due to a given source. Here, we partitioned indoor and personal exposure concentrations to seven c-PAHs and pyrene according to the indoor- and outdoor-origins.
Methods: A simultaneous, integrated monitoring of personal, indoor and outdoor concentrations of nine PAHs was conducted in 75 homes for a consecutive 48-hour period across a two-year period in Kraków, Poland. Due to few known indoor sources for chrysene, we used this PAH species as a tracer for infiltration of outdoor PAHs. Personal and indoor concentrations of seven c-PAHs and pyrene were apportioned to home indoor, non-home indoor and outdoor origins.
Results: Using $Chrysene_{in}/Chrysene_{out}$ as proxy for an infiltration factor, F_{inf}, infiltrated PAHs of outdoor origin are overall higher in concentration than those emitted from the indoor origin. Average contribution by the outdoor sources on B[*a*]A, B[*b*]F, and B[*k*]F were 92%, 79%, and 78% across all seasons, respectively. In contrast, in homes where a household member smoked, average contributions by the outdoor sources on B[*ghi*]P, B[*a*]P, D[*ah*]A, and IP were lower (i.e., 67%, 65%, 67%, and 66%, respectively). Season-averaged contributions by the outdoor sources on personal exposure to B[*a*]A, B[*b*]F, and B[*k*]F were 92%, 74%, and 77%, respectively. On the other hand, season-averaged home indoor source contributions on personal exposure to B[*a*]A, B[*b*]F, and B[*k*]F were estimated at 6%, 15%, and 19%, respectively. Similar contributions by season-averaged home indoor sources on personal exposure were estimated at 28% for B[*ghi*]P, 31% for B[*a*]P, 25% for D[*ah*]A, and 28% for IP.
Conclusion: Of the seven c-PAHs, B[*a*]A, B[*b*]F, and B[*k*]F are enriched in indoor and personal exposure concentrations from the outdoor coal-combustion. B[*ghi*]P, B[*a*]P, D[*a,h*]A, and IP, PAHs with some of the highest carcinogenic and mutagenic potencies, are considerably enriched by cigarette smoke in addition to the outdoor sources.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) constitute particle-bound or gaseous component of indoor smoke with carcinogenic and a number of other effects on human health (WHO, 2000a, 2000b). Yet, understanding the health risks from indoor exposure to airborne PAHs faces a set of unique challenges. Such challenges include scarcity of information regarding the indoor and outdoor sources as well as the conditions which mediate human exposure (e.g. fuel type, housing age, housing structural material, ventilation quality, and food preparation method) (Smith and Mehta, 2003). Specifically, variable rates of environmental photodegradation of the PAHs, human behavior (e.g., cooking, cigarette smoking, cleaning), weather conditions (e.g., temperature, relative humidity, wind speed), characteristics of the residential building (Spengler et al., 1996), and indoor environment conditions (e.g., ventilation, temperature, ultraviolet ray, humidity) could modify the primarily generated PAHs through removal or secondary generation (Schauer et al., 2003). Apart from such chemical interactions, the toxicity of PAH mixture within in vitro models has been shown to be synergistic or antagonistic compared to those of component individual compounds (Tarantini et al., 2011).

To date, the extent to which multiple indoor and outdoor sources contribute to personal PAH exposure concentration remains very poorly

Abbreviations: PAHs, Polycyclic aromatic hydrocarbons; c-PAHs, carcinogenic PAHs; B[*a*]A, benz[*a*]anthracene; B[*a*]P, benzo[*a*]pyrene; B[*b*]F, benzo[*b*]fluoranthene; B[*k*]F, benzo[*k*]fluoranthene; B[*ghi*]P, benzo[*ghi*]perylene; EF, Enrichment factor; IP, indeno[*123-cd*]pyrene; D[*ah*]A, dibenz[*ah*]anthracene; I/O, Indoor/outdoor; SHS, Secondhand smoke; MDRs, Molecular diagnostic ratios; HDD, heating degree days; RTEF, reference toxic equivalency factor; TEF, toxic equivalency factor.

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understood. Popular approach to estimating personal exposure includes using centrally located ambient monitor. One of the essential assumptions of such approach is that the ambient pollutant concentration is highly correlated with the outdoor-originating portion of the personal exposure (Wallace and Williams, 2005). Implicitly, such assumption postulates that personal exposure to outdoor originating PAHs significantly contributes to health outcomes. However, as adults spend an estimated 80 to ≥90% of daily hours within indoor microenvironments (Brunekreef et al., 2005; Samet and Spengler, 2003), health consequences from personal exposure to indoor-originating PAHs may often be underestimated. For example, the PAH mixture emitted from cigarette smoke is estimated to possess higher carcinogenic potencies compared to that emitted from diesel engine exhaust (Valberg and Watson, 1999). Furthermore, reliance on coal-burning for home heating during early life is associated with impaired skeletal growth at the 36th month of age (Ghosh et al., 2011).

Based on such evidence, estimating personal exposure contribution by outdoor-originating PAHs represents an important research need. In this investigation, we follow our previous inquiry (Choi et al., 2008a), to quantify the extent to which indoor and outdoor sources influence the indoor and personal exposure concentrations of eight PAHs. Earlier source apportionment and economic analyses have shown that airborne PAHs in Krakow are predominantly generated from coal-burning in low-efficiency residential stoves and boilers (Junninen et al., 2009; Lvovsky et al., 2000). In addition, we showed that pregnant women are exposed to a sharp seasonal trend in infiltrated PAHs (Choi et al., 2008a). Based our earlier recognition of outdoor-originating PAHs and secondhand cigarette smoke as two important sources (Choi et al., 2008a), here, we quantify relative contributions of such sources on personal exposure.

We conducted such analyses by adapting the sulfur tracer method (Sarnat et al., 2009; Wallace and Williams, 2005) and enrichment factor to meet following specific aims: 1) partition the indoor PAH concentration according to the indoor and outdoor sources; 2) estimate the contribution of the outdoor versus indoor-based PAHs in personal exposure concentration; and 3) explore the utility of enrichment factor as predictive marker of the proximity effect in personal exposure cloud.

2. Methods

Details on the subject enrollment and air monitoring methods have been published (Choi et al., 2006; Jedrychowski et al., 2004, 2006) and briefly summarized below.

2.1. Study site characterization

The city of Kraków in south of Poland represents one of the areas in Europe with historically intensive coal-burning power generation (Junninen et al., 2009). Additional sources of local air pollution include commercial activities with high automobile traffic within the Kraków city center, and coal-burning for home heating (Junninen et al., 2009). Women in the cohort study live in the urbanized area of Kraków (Jedrychowski et al., 2004). The easternmost district of Kraków, Nowa Huta, encompasses several steel mills, including an iron ore sinter plant, blast furnace, coke, gas and coal combustion power plant, natural gas-fired steel production plant, and oxygen furnace steel plant. The same district also contains a coal-fired cement kiln, and a coal-fired power plant (Junninen et al., 2009).

2.2. Subject enrollment

Briefly, we recruited young (aged 18–35), non-smoking pregnant women with no known pre-existing risks of adverse birth outcomes from the prenatal care clinics throughout the seasons (23% December–February, 27% March–May, 27% June–August, and 24% September–November) in Kraków (n = 344). During the late 2nd trimester, a research questionnaire was administered and collected information on demographic and socioeconomic status as well as description of the surrounding outdoor environment. The questionnaire inquired about indoor features, active and passive smoking, dietary intake of PAH-containing foods, as well as other daily activities. Passive smoking was self-reported in terms of duration (hours/day) and intensity (number smoked/day). The institutional review board of Columbia Presbyterian Medical Center approved the study, and informed consent was obtained from all study participants.

2.3. Indoor and outdoor air monitoring

As described elsewhere (Choi et al., 2008a, 2008b), we conducted the home indoor and the home outdoor PAH monitoring simultaneously for a 48-hour period using two identical monitors. Briefly, we fitted a backpack with the URG-2000-25 Personal Air Sampler (URG, Chapel Hill, NC, USA). The impactor inlet was fastened to the top of the shoulder strap, to collect air sample near the woman's breathing zone. For the indoor measurement, we placed an identical backpack in a room where the woman spent most of her time at home (i.e. living room, bedroom or near the kitchen). The sampler was placed atop a furniture 0.5–2 m above the floor away from the heating source or the window. For the outdoor measurement, we secured an identical monitor at a window height usually in the balcony, about one meter away from the wall of the home or the apartment. The sampling pump (BGI, Waltham, MA, USA) with the split flow inlet drew in the air continuously at 2 l/min. The pump flow was split two ways to simultaneously collect particles ≤2.5 μm in aerodynamic diameter ($PM_{2.5}$) and PAHs. PAHs were collected on a quartz microfiber filter (Palliflex Tissuquartz 2500 QAS, 25 mm in diameter) and semi-volatile vapors and aerosols were collected on a polyurethane foam (PUF) plug backup (Kinney et al., 2002). Personal air monitoring data were given a Quality Assurance (QA) score (0–3) for flow rate, flow time, and completeness of documentation (Kinney et al., 2002). Most samples were shipped to the laboratory within 60 days of sample collection, and were extracted within 14 days after arrival.

2.4. Statistical analyses

We limited our analysis to the indoor and outdoor air samples with a high/good quality assurance (QA) score (0 or 1), which resulted in 76 (100%) personal, 76 (97%) indoor, and 76 (91%) outdoor samples. The indoor/outdoor ratios of the nine PAHs were calculated for 75 households with simultaneous measurements. The indoor and outdoor exposure levels of nine individual PAHs were skewed (all *p*-values for Kolmogorov–Smirnov test <0.001). After natural log (ln) transformation, the distribution of the indoor and outdoor measurements conformed to normal distribution. There were no PAH concentrations below the detection limit. Season of PAH monitoring is defined as summer (June–August), transition (March–June and September–November), and winter (December–February).

In our earlier analyses, an indicator of secondhand smoke at home was the only significant indoor source of personal exposure (Choi et al., 2008a). We also validated secondhand smoke exposure using cotinine (Choi et al., 2006). That is, both maternal and newborn levels were within the range expected for the secondhand smoke exposure (Choi et al., 2006).

Here, we analyzed secondhand smoke as hours/day of exposure as well as the number smoked in the presence of the pregnant women.

2.4.1. Contribution of indoor PAHs by indoor and outdoor sources

The indoor concentrations of the eight PAHs were estimated by adapting a sulfur tracer method (Sarnat et al., 2002, 2009; Wallace and Williams, 2005). Since we did not measure the factors such as particle infiltration, exfiltration, deposition, air exchange rate, the corresponding gas–particle partition kinetics, particle penetration, and particle deposition, we investigated chrysene as a tracer PAH compound to