Zdenek Fiala · Adolf Vyskocil · Vladimir Krajak
Claude Viau · Eva Etterlova · Josef Bukac
Dana Fialova · Stanislav Emminger

Environmental exposure of small children to polycyclic aromatic hydrocarbons

Received: 18 September 2000 / Accepted: 20 February 2001

Abstract Objectives: The aim of the study was to assess the intake (by various routes of exposure) of polycyclic aromatic hydrocarbons (PAH) by children living in a Czech city, and its effect on excretion of 1-hydroxypyrrene (1-OHP) in summer and winter periods. Methods: Four groups of children (3–6 years old) were chosen: (1) two groups from a kindergarten situated in the city center with a higher traffic density (“polluted” area); (2) two groups from a kindergarten situated in a green zone of the same city (“non-polluted” area). Food consumption was recorded in all children and PAH intake from foodstuffs was estimated. Ambient air samples were collected from the playground and inside the kindergartens. Soil samples were collected too. Morning and evening urine samples were collected during sampling days. Results: In both seasons, the mean outdoor total PAH concentration (sum of 12 individual PAH) in the “polluted” area was approximately three-times higher than that in the “non-polluted” area. Indoor concentration in the “polluted” area was more than six-times higher than that in the “non-polluted” area in summer, and almost three-times higher in winter. The same trend was observed for pyrene and for the sum of carcinogenic PAH. The contribution to the total pyrene absorbed dose from food consumption was much more important than that from inhalation and from ingestion of soil dust. Significantly higher urinary concentrations of 1-OHP (evening samples) were found in children from the “polluted” kindergarten in both seasons. The number of significant relationships between 1-OHP and pyrene absorbed dose was weak. Conclusions: Food seems to be the main source of total pyrene and total PAH intake in small children, even under relatively higher air PAH exposure in the city. Estimated pyrene ingestion from soil had a negligible contribution to the total pyrene absorbed dose. Urinary 1-OHP seems to be an uncertain (non-sensitive) marker of the environmental inhalation exposure to pyrene (PAH) if the pollution of air by pyrene (PAH) is not excessive and the pyrene (PAH) dose by this route is much less than by ingestion. Usefulness of the urinary 1-OHP as an indicator of overall environmental exposure to PAH needs further investigation.

Key words Polycyclic aromatic hydrocarbons · Children · Environmental exposure · 1-Hydroxypyrrene

Introduction

Polycyclic aromatic hydrocarbons (PAH) are now well-established as human health hazards. A number of these are known to have carcinogenic and mutagenic properties. Human exposure to PAH occurs principally by (1) direct inhalation of polluted air and tobacco smoke, (2) dietary intake of smoked and other foodstuffs and polluted water, and (3) dermal contact with soot, tar, and polluted soils (International Agency for Research on Cancer (IARC) 1983).

The assessment of environmental exposure to PAH requires the measurement of levels present in each
pathway of possible contact. The biological monitoring of PAH represents the best way to assess the internal dose of PAH. Among the many PAH, pyrene is emitted in fairly large amounts. Pyrene is contained not only in exhaust from heaters, but also in automotive exhaust, cigarette smoke, and soil, as well as in food (Jongeneelen, 1994). Pyrene is metabolized and excreted as 1-hydroxypyrene (1-OHP) in urine in an amount dependent on the dose of pyrene. It was suggested that 1-OHP is a useful marker in exposure evaluation to pyrene, thus to PAH in general, as well as in assessment of their health effects. Indeed, several industrial hygiene studies have shown that 1-OHP in urine is a valid biomarker for occupational exposure to PAH (Buchet et al. 1992; Jongeneelen et al. 1990; Zhao et al. 1990).

Currently, urinary 1-OHP as well as selected hydroxylated metabolites of phenanthrene are tested as biomarkers for the assessment of low-level environmental exposure of general population (Gündel et al. 1996; Seiwert et al. 1998; Viau et al. 1995). Our latest reports (Gilbert and Viau 1997; Vyskocil et al. 1997, 2000) and reports by Kanoh et al. (1993), Jongeneelen (1994) and Zhao et al. (1992) suggest that levels of 1-OHP reflect the levels of environmental PAH exposure from different sources such as air, food and the environment of the home life. However, in our last studies (Vyskocil et al. 1997, 2000), it was shown that factors other than air pollution contribute more substantially to overall exposure to PAH, and it was speculated that diet could be one of the most important factors.

Up to now, only one study (Van Wijnen et al. 1996) followed up the intake of PAH in small children, who could be more exposed to PAH than adults. It is known that small children spend much of their time outdoors in close contact with soil. So, in children, dermal contact with soil polluted by PAH, ingestion of soil and inhalation of soil particles could be further routes for PAH intake, that are not so important in adults. PAH from vehicle exhausts and road runoff have been suggested as major sources of contamination of the roadside environment (Pathirana et al. 1994). This could present an important source of PAH exposure for children playing in the vicinity of roads.

The objective of our investigation was to assess the multi-pathway intake of PAH by small children living in a city. More specifically, it was designed to (1) determine concentrations of individual PAH in air and soil of a highly and a less polluted area of the city of Hradec Králové (Czech Republic) where they are exposed; (2) estimate intake of PAH from the diet on the basis of a detailed questionnaire; (3) identify among the multiple sources (air, soil, diet) relative magnitudes of exposure to PAH in children; (4) determine the effect of the PAH intake on 1-OHP concentrations in urine.

**Methods**

Four groups of children (3–6 years old) living in the city of Hradec Králové were chosen. Two groups (n = 15 in summer and n = 10 in winter) were chosen from a kindergarten (identified as a “polluted area”) situated near an important road (the central part of the playground was situated at a distance of 22 m from the road) with high traffic density (about 18,000 cars/day). Two groups (n = 15 in summer and n = 17 in winter) were chosen from a kindergarten (identified as a “non-polluted area”) situated in a residential zone with low local traffic (about 80 cars/day). All children lived in the vicinity of the corresponding kindergartens, in houses situated on small streets with local circulation only. Informed consent was obtained from the parents of children prior to their inclusion in the study, and the Ethics Committee of Charles University Medical Faculty in Hradec Králové approved the work.

One day before, and during sampling periods, detailed food consumption was recorded in all subjects by parents and teachers. During the sampling period, the time that the children spent outside and inside the kindergarten was recorded. The parents answered several questions concerning the other possible sources of PAH exposure in children (smoking habits of parents in the home, heating of houses, cooking devices in houses, frequency of grilled, baked, smoked and roasted food and other possible sources of PAH exposure in the residential area).

Daily pyrene intake from foodstuffs was estimated using a method described in our previous publication (Chen et al. and Viau 1997). In brief, using literature data, we drew up a table where foods were divided into four classes according to their pyrene concentrations: low (<1 μg/kg, taken as 0.5 μg/kg), medium (1–5 μg/kg, taken as 3 μg/kg), high (5–25 μg/kg), taken as 15 μg/kg) and very high (>25 μg/kg, taken as 75 μg/kg). A description of every meal was recorded in a diary, and the pyrene content in all foods was calculated.

Ambient air samples were collected in the playgrounds (the sampler was placed in the center of the playground) and inside the kindergartens (the sampler was placed in the central corridor) for three consecutive days in summer 1997 (three samples per site) and winter 1998 (three samples per site). The collection of ambient air by stationary air samplers (duration of sampling = 8–14 h; air flow =20 m³/h) and its analysis for PAH were performed by the EPA Toxic Method (Environmental Protection Agency (EPA) 1988). The air samples were collected on quartz-fiber filters and polyurethane foams (PUF). Quartz-fiber filters were extracted with methylene chloride, and PUF plugs with 10% diethylether in hexane in a Soxhlet apparatus. The extracts were dried by being passed through the column containing anhydrous sodium sulphate, and were concentrated in a rotary evaporator. The concentrated extracts were cleaned by being passed through a silica gel column. Gradient high-performance liquid chromatography (HPLC) with fluorescence detection was used for qualitative and quantitative determination of individual PAH. In each sample the concentration of 12 selected PAH compounds (phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, diben[a,h]anthracene, benzo[ghi]perylene, and indeno[1,2,3-cd]pyrene) was determined. Total PAH concentration was calculated as the sum of the concentrations of the 12 PAH.

Atmospheric conditions during summer collections were partially sunny. The temperatures ranged from 14.7 to 22.1 °C and the wind blew at 6–9 m/s. During winter collections we observed partially sunny conditions as well, with temperatures from −0.3 to −4.2 °C and with the wind from 6 to 14 m/s.

The urine samples were collected during the 3 days that the air samples were collected, in the morning and in the evening (before the child went to sleep). Urine samples were analyzed for 1-OHP by a gradient HPLC method with fluorescence detection, that was adapted in our laboratory (Bouchard et al. 1994). Urine creatinine was used to standardize the results.

Soil samples were taken from the upper soil layer (first 20 cm). Each representative sample of soil was prepared from a mixture of samples collected from five different parts of the playgrounds and sandpits in each kindergarten. In winter the children did not play in the sandpits. For this reason we did not collect the sand samples then. All soil samples were analyzed for PAH by a modified HPLC method according to Haak and Ham (1994). The samples were