Indoor concentrations of fine particles and particle-bound PAHs in Gothenburg, Sweden

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Abstract. Fine particles are formed in a variety of processes, both natural and anthropogenic. Epidemiological studies have shown an association between exposure to particulate matter and adverse health effects. Airborne particles contain a variety of compounds, including polycyclic aromatic hydrocarbons (PAHs), and several of the PAHs are known or suspected carcinogens. In this study, stationary measurements of PM2.5 were performed in the residences of 20 study participants along with simultaneous monitoring at an urban background site. The collected particle mass was then analyzed for its content of some particle-bound PAHs using GC-MS. The median level of PM2.5 indoors was 7.3 µg/m3 and in urban background 5.3 µg/m3. For benzo(a)pyrene (B(a)P) the corresponding results were 10 pg/m3 and 35 pg/m3, respectively. There were significant correlations between indoor and ambient levels for both PM2.5 (rs=0.58, p=0.02) and B(a)P (rs=0.67, p=0.007). No significant correlation was, however, found between the concentration of PM2.5 and the associated levels of the investigated PAH compounds. This finding implies that exposure to B(a)P or other particle-bound PAH components needs to be separately assessed.

1. Introduction

Fine particles are formed in a variety of processes, both natural and anthropogenic. Traffic, burning of biomass and other fuels for heating purposes and industry are important sources of fine particles in urban areas. Epidemiological studies have shown an association between exposure to particulate matter and adverse health effects such as cancer, cardiovascular diseases and respiratory illnesses [1, 2]. In southern Sweden, long-range transported particles give a substantial contribution to ambient levels.

Airborne particulate matter contains a variety of compounds, including polycyclic aromatic hydrocarbons (PAHs), which are formed during incomplete combustion processes. Domestic wood-burning and traffic are the two main contributors to the release of PAHs in Sweden [3]. Several known or suspected carcinogens are found among the PAHs, and many of these are 5- and 6-ring compounds that are associated with particles. Benzo(a)pyrene (B(a)P) is the most studied among the carcinogenic PAHs. According to the unit risk estimate from the WHO [4], the exposure to B(a)P that will cause an excess lifetime cancer risk of 1/100 000, is 0.1 ng/m3. This level is used as a Swedish health based guideline value [3].

Within the frame work of the Swedish environmental protection agency, a project concerning the general population’s exposure to carcinogenic compounds was started in the year 2000. The project includes five Swedish cities, one of them being Gothenburg. The health related survey includes...
measurements of benzene, 1,3-butadiene, formaldehyde and nitrogen dioxide. Measurements of fine particles were added to the project in 2005. This study presents results from the measurements of PM$_{2.5}$ and some particle-associated PAHs in Gothenburg during the fall 2006.

2. Materials and methods

2.1. Study group
Adult citizens (age 20-50 years) living in Gothenburg were randomly selected from the population register. Among 40 study participants, 10 individuals were randomly selected for home indoor measurements of fine particles. In addition to these 10 individuals, another 10 volunteers were recruited among the employees at the Department of Occupational and Environmental Medicine in Gothenburg.

2.2. Monitoring
Stationary measurements of fine particles (PM$_{2.5}$) were performed indoors (living room) in the homes of the 20 study participants during the fall of 2006. The sampling was performed for 48 hours using GK2.05 (KTL) cyclones connected to BG1400S sampling pumps (BGI Inc., Waltham, MA, USA) with a flow rate of 4 L/min. The flow rate was adjusted prior to monitoring and controlled at the end of the sampling period. The average flow rate was then used to calculate the total volume of air drawn through the filter. Simultaneously with the indoor sampling (for 17 of the 20 subjects), urban background monitoring was conducted at a central site within the city using a PQ100 Basel PM$_{2.5}$ sampler with an impactor cutoff system (EPA WINS, BGI) at 16.7 L/min. On four occasions, indoor sampling was performed simultaneously in the homes of two study subjects. Consequently, on these occasions two indoor samples were accompanied by a single urban background sample (one sample missing due to instrument failure).

2.3. Analyses
Teflon filters, 2 µm pore size (Pall Teflo) were used for all measurements. All filters were weighed before and after exposure. The filters were conditioned for 24 hours prior to weighing in a climate chamber controlled for temperature and humidity (temperature: 23 ±0.5°C, relative humidity (RH): 50±5 %). The mass concentration was determined using a Mettler Toledo MX5, and the limit of detection was 1 µg/m$^3$.

The particle mass was then analyzed for its content of particulate PAHs. Analysis and detection was performed using high-resolution gas chromatography (GC) connected to a low-resolution mass spectrometer (MS) in selective ion recording (SIR) mode. Field blanks were processed in parallel with the samples. An internal standard (16 PAHs) was used in the analysis. A reference material (SRM 1649a urban dust) certified for 12 PAH components was used as control samples. The results for these control samples showed acceptable agreement with the certified results for all components. The detection limit was 5 pg/m$^3$ for the indoor samples, and 1 pg/m$^3$ for the urban background samples.

2.4. Statistics
Statistical calculations were performed with the SAS System for Windows, version 9.1 (2003). Correlations were assessed using the Spearman rank correlation coefficient ($r_s$) and differences between pairs of indoor and ambient levels were calculated using the Wilcoxon signed rank test. Statistically significant refers to 5 % significance level in two-tailed tests ($p<0.05$). If the particle mass was below the LOD, the LOD divided by the square root of 2 was used [5].
3. Results

In total, the particle mass was analyzed for nine different PAHs. For three of the components, benzo(a)anthracene, perylene and dibenzo(a,h)anthracene, more than 50 % of the indoor samples were below the detection limit. The median concentration, mean and range for the indoor and urban background measurements of PM$_{2.5}$ and the six detectable PAH components are listed in table 1.

| Compound                   | Indoor                        | Urban background          |
|----------------------------|-------------------------------|---------------------------|
|                            | N    | Median (pg/m$^3$) | Mean (pg/m$^3$) | Range (pg/m$^3$) | N    | Median (pg/m$^3$) | Mean (pg/m$^3$) | Range (pg/m$^3$) |
| PM$_{2.5}$ (µg/m$^3$)      | 20   | 7.3            | 9.7              | 3.2-45         | 11   | 5.3            | 7.3              | 2.9-18          |
| Chrysene                   | 20   | 6             | 23               | <5-290         | 11   | 30            | 39               | 30-110          |
| Benzo(b)fluoranthene       | 20   | 15            | 40               | <5-370         | 11   | 72            | 83               | 26-280          |
| Benzo(k)fluoranthene       | 20   | 7             | 27               | <5-340         | 11   | 67            | 85               | 10-230          |
| Benzo(a)pyrene             | 20   | 10            | 32               | <5-340         | 11   | 35            | 46               | 11-140          |
| Indeno(1,2,3-c,d)pyrene    | 20   | 41            | 69               | <5-390         | 11   | 83            | 91               | 28-280          |
| Benzo(g,h,i)perylene      | 20   | 37            | 73               | <5-390         | 11   | 85            | 95               | 28-260          |

3.1. Particle mass

The median concentrations from the parallel measurements of PM$_{2.5}$ indoors in living rooms and in urban background are presented in figure 1. The median concentration of PM$_{2.5}$ indoors was 7.3 µg/m$^3$ (95 % CI 5.9-9.3 µg/m$^3$) and in urban background 5.3 µg/m$^3$ (95 % CI 3.3-17 µg/m$^3$). The highest level of PM$_{2.5}$ was found in the home of the one smoker (subject 9) who also smoked indoors.

![Figure 1](image.png)

**Figure 1.** Median levels of PM$_{2.5}$ indoors and in urban background, respectively. Subject 1, 7 and 9 were smokers, but only subject 9 smoked indoors.

3.2. Benzo(a)pyrene

Results from the parallel indoor and urban background measurements of B(a)P are shown in figure 2. The median B(a)P concentrations indoors and in urban background were 10 pg/m$^3$ (95 % CI 5-23 pg/m$^3$) and 35 pg/m$^3$ (95 % CI 13-110 pg/m$^3$), respectively. Also for B(a)P, the highest concentration was found in the home of the smoker who also smoked indoors.
3.3. Relations between concentrations in different microenvironments

There was a statistically significant correlation between indoor and ambient concentrations for both PM$_{2.5}$ (N=15; r$_s$=0.58, p=0.02) and B(a)P, (N=15; r$_s$=0.67, p=0.007). Omitting the one smoker who also smoked indoors (subject 9) from the dataset resulted in a nearly significant correlation for PM$_{2.5}$ and a slightly reduced, but still significant correlation coefficient for B(a)P, see figure 3 (a) and (b).

A significant correlation between indoor and urban background levels was not only shown for B(a)P, but also for the other compounds (r$_s$=0.66-0.84). Urban background concentrations were, however, significantly higher than indoor levels for all the investigated PAHs. In general, the PAH
components correlated well with each other, which holds for both the indoor and outdoor measurements. There was, however, no correlation between particle mass concentrations and the different PAH components for neither the indoor measurements, nor at the urban background site.

4. Discussion

Indoor concentrations of PM$_{2.5}$ were low compared to levels found in studies conducted in central and southern Europe [6], but comparable to levels found in Helsinki [7] and a previous study in Gothenburg [8]. Also the measured levels of B(a)P must be considered low. However, Clayton et al. [9] found comparable indoor B(a)P levels in a study conducted in Minnesota, while most of the concentrations measured in the study from United States by Naumova et al. [10] were higher. Also the study from Berlin [11] reported higher median levels of B(a)P compared with Gothenburg.

The method for analysis of the PAHs in this study is the same as in the study by Pleil et al.[13], where PM$_{2.5}$ filters were analyzed for their content of PAHs. In that study, a mean concentration of B(a)P of 87 pg/m$^3$ was reported from outdoor measurements in North Carolina. In another Swedish study, in an area where residential wood-burning is common, elevated indoor levels of several PAHs were found in the residences of study subjects who used wood-burning appliances for space heating [12]. In that study, median indoor B(a)P concentrations of 520 pg/m$^3$ was found in the wood-burning homes and 120 pg/m$^3$ in the reference homes. In the present study from Gothenburg, however, none of the study participants used wood for space heating.

PAHs with 3-4 rings are classified as semi-volatile and can have a significant fraction in both the gaseous and particulate phases [3, 12]. This might have contributed to the quite low levels of chrysene detected in the collected particle mass. However, the remaining five detected PAH components are 5-6 ring compounds that are all found in the particulate phase [3, 13].

No correlation was found between PM$_{2.5}$ and levels of B(a)P or the other individual PAH components. This is in consistence with the findings by Ohura et al. [14]. In the study by Fromme et al. [11], no correlation was found between particle mass and a majority of the investigated PAHs, apart for measurements conducted in smoker households. The finding of no association between particle mass and PAHs implies that exposure to B(a)P or other particle-associated PAHs needs to be characterized separately. Finally, in the present study we found the median levels of PM$_{2.5}$, both indoors and at the urban background site, to be below the WHO guideline value of 10 µg/m$^3$ [15]. Furthermore, the median concentration of B(a)P was well below the Swedish health based guideline value.

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References

[1] Pope CA and Dockery DW. Health effects of fine particulate air pollution: lines that connect. J Air Waste Manag Assoc. 2006 Jun;56(6):709-42.
[2] Schlesinger RB, Kunzli N, Hidy GM, Gotschi T and Jerrett M. The health relevance of ambient particulate matter characteristics: coherence of toxicological and epidemiological inferences. Inhal Toxicol. 2006 Feb;18(2):95-125.
[3] Bostrom CE, Gerde P, Hanberg A, Jernstrom B, Johansson C, Kyrklund T, Rannung A, Törnqvist M, Victorin K and Westerholm R. Cancer risk assessment, indicators, and guidelines for polycyclic aromatic hydrocarbons in the ambient air. Environ Health Perspect. 2002 Jun;110 Suppl 3:451-88.
[4] WHO. Air Quality Guidelines for Europe. 2nd Edition ed: WHO 2000.
[5] Hornung RW and Reed LD. Estimation of average concentration in the presence of nondetectable values. Applied Occupational and Environmental Hygiene. 1990;5(1):46-51.
[6] Gotschi T, Oglesby L, Mathys P, Monn C, Manalis N, Koistinen K, Jantunen M, Hanninen O, Polanska L and Künzli N. Comparison of black smoke and PM2.5 levels in indoor and outdoor environments of four European cities. *Environ Sci Technol*. 2002 Mar 15;36(6):1191-7.

[7] Koistinen KJ, Hanninen O, Rotko T, Edwards RD, Moschandreas D and Jantunen MJ. Behavioral and environmental determinants of personal exposures to PM2.5 in EXPOLIS - Helsinki, Finland. *Atmospheric Environment*. 2001;35(14):2473-81.

[8] Johannesson S, Gustafson P, Molnar P, Barregard L and Sallsten G. Exposure to fine particles (PM2.5 and PM1) and black smoke in the general population: personal, indoor, and outdoor levels. *J Expo Sci Environ Epidemiol*. 2007 Nov;17(7):613-24.

[9] Andrew Clayton C, Pellizzari ED, Whitmore RW, Quackenboss JJ, Adgate J and Sefton K. Distributions, associations, and partial aggregate exposure of pesticides and polynuclear aromatic hydrocarbons in the Minnesota Children's Pesticide Exposure Study (MNCPES). *J Expo Anal Environ Epidemiol*. 2003 Mar;13(2):100-11.

[10] Naumova YY, Eisenreich SJ, Turpin BJ, Weisel CP, Morandi MT, Colome SD, Totten L, Stock T, Winer A, Alimokhtari S et al. Polycyclic aromatic hydrocarbons in the indoor and outdoor air of three cities in the U.S. *Environ Sci Technol*. 2002 Jun 15;36(12):2552-9.

[11] Fromme H, Lahrz T, Piloty M, Gebhardt H, Oddoy A and Ruden H. Polycyclic aromatic hydrocarbons inside and outside of apartments in an urban area. *Sci Total Environ*. 2004 Jun 29;326(1-3):143-9.

[12] Gustafson P, Östman C and Sällsten G. Indoor levels of polycyclic aromatic hydrocarbons in homes with or without wood burning for heating. *Environ Sci Technol*. 2008(DOI: 10.1021/es800304y).

[13] Pleil JD, Vette AF and Rappaport SM. Assaying particle-bound polycyclic aromatic hydrocarbons from archived PM2.5 filters. *J Chromatogr A*. 2004 Apr 9;1033(1):9-17.

[14] Ohura T, Amagai T, Sugiyama T, Fusaya M and Matsushita H. Characteristics of particle matter and associated polycyclic aromatic hydrocarbons in indoor and outdoor air in two cities in Shizuoka, Japan. *Atmospheric Environment*. 2004;38(14):2045-54.

[15] WHO. *WHO Air Quality Guidelines Global Update 2005*. Copenhagen, Denmark: World Health Organization, Regional Office for Europe, Bonn, Germany.; 2005.