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Submicron particle-bound polycyclic aromatic hydrocarbons in the Polish teaching rooms: Concentrations, origin and health hazard

Wioletta Rogula-Kozłowska[1,2,*], *Barbara Kozielska*[3], *Grzegorz Majewski*[4], *Patrycja Rogula-Kopiec*[1], *Walter Mucha*[3], *Karolina Kociszewska*[4]

1. *Institute of Environmental Engineering, Polish Academy of Sciences, 34 M. Skłodowska-Curie St., 41-819 Zabrze, Poland*
2. *The Main School of Fire Service, Faculty of Fire Safety Engineering, 52/54 Słowackiego St., 01-629 Warsaw, Poland*
3. *Silesian University of Technology, Faculty of Energy and Environmental Engineering, Department of Air Protection, 22B Konarskiego St., 44-100 Gliwice, Poland*
4. *Faculty of Civil and Environmental Engineering, Warsaw University of Life Sciences, 166 Nowoursynowska St., 02-776 Warsaw, Poland*

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ABSTRACT

The goal of the work was to investigate the concentrations of the 16 US EPA priority polycyclic aromatic hydrocarbons (PAH) bound to submicrometer particles (particulate matter, PM_1) suspended in the air of university teaching rooms and in the atmospheric air outside. Two teaching rooms were selected in two Polish cities, Gliwice, southern Poland, and Warsaw, central Poland, differing with regard to the ambient concentrations and major sources of PM and PAH. The variabilities of indoor and outdoor 24-hr concentrations of PM_1-bound PAH, the ratio (I/O) of the indoor to outdoor 24-hr concentrations of PM_1-bound PAH, probable sources of PAH and the level of the hazard from the mixture of the 16 PAH (ΣPAH) to humans at both sites were analyzed. In both Warsaw and Gliwice, the mean concentrations of PM_1-bound ΣPAH were slightly higher in the atmospheric air than in the rooms. The indoor and outdoor concentrations of individual PAH in Gliwice were correlated, in Warsaw – they were not. Most probably, the lack of the correlations in Warsaw was due to the existence of an unidentified indoor source of gaseous PAH enriching PM_1 in phenanthrene, fluorene, and pyrene. Although the ambient concentrations of PM_1-bound PAH were low compared to the ones observed earlier at both sites, they were much higher than in other urbanized European areas. However, because of low mass share of heavy PAH in ΣPAH, the various indicators of the health hazard from the 16 PAH mixture were low compared to other regions.

Introduction

In general, the air quality inside buildings depends on the atmospheric air quality. This dependency can be questionable for hermetically sealed rooms, where air is exchanged through ventilation or air conditioning systems, but if there are neither forced air exchange nor inside pollution sources, even air tight doors and windows do not prevent the pollutant concentrations in the room and in the atmospheric air outside from being very close. In hot climate zones, where the windows and doors are

* *Corresponding author*. E-mail: wioletta.rogula-kozlowska@ipis.zabrze.pl, wrogula@sgsp.edu.pl (Wioletta Rogula-Kozłowska).

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permanently open, this dependency is obvious, even when inside pollution sources exist (Jones, 1999; Monn, 2001; Morawska and Salthammer, 2003).

The closed doors and windows in normal buildings practically are not an obstacle to ambient fine particulate matter (fine PM: $PM_{2.5}$, PM_1) getting inside (Jones et al., 2000; Morawska and Salthammer, 2003). In public facilities (schools, nurseries, offices, libraries), if there are no indoor pollution sources, the indoor fine PM concentrations are close to (or slightly higher than) the PM concentrations outside. (*e.g.*, Hassanvand et al., 2015; Romagnoli et al., 2014; Oliveira et al., 2016; Krugly et al., 2014; Zhu et al., 2015; Alves et al., 2014; Błaszczyk et al., 2017). However, because polycyclic aromatic hydrocarbons (PAH) adhere to PM particles only in some specific conditions, the indoor concentrations of PM-bound PAH can be visibly lower than outdoor ones (ibid.). The mass proportions of the PM-bound and volatile phases of PAH, especially of 2-, 3-, and 4-ring ones, are not stable (Tobiszewski and Namieśnik, 2012; Keyte et al., 2013). They vary with varying ambient concentration and chemical composition of PM, ambient concentrations of co-occurring substances, temperature and relative humidity of air, *etc.* (Ravindra et al., 2008; Rybak and Olejniczak, 2014).

PAH are considered one of the most hazardous components of PM. They are the first air pollutants identified as carcinogens by the US Environmental Protection Agency (US EPA). The strength of their carcinogenicity grows with their molecular weight. After entering the human body, PAH are conveyed by blood to various organs, where they are metabolized (Nikolao et al., 1984; White, 2002; Keyte et al., 2013). The metabolites, after they covalently bond with DNA or RNA, cause neoplasms, affect replications, transcriptions, and biosynthesis of proteins (Skupińska et al., 2009). All PAH can accumulate in the body tissues. However, it is unknown so far if the carcinogenicity of PAH, which in the air occur always as a mixture, never as a single compound, may be ascribed to individual hydrocarbons or if it is due to the concerted effects of some number of PAH. The carcinogenicity of a PM-bound PAH may be enhanced or suppressed by other PM components.

PM_1, the core part of PM in urbanized areas, contains majority of PM-bound PAH (*e.g.*, Rogula-Kozłowska, 2015, 2016). In Poland, especially in the urbanized areas of its southern part, ambient concentrations of PM- and PM_1-bound PAH are highest in Europe (Rogula-Kozłowska et al., 2013; Rogula-Kozłowska, 2015). The indicator of PM-bound PAH air pollution, PM_{10}-bound benzo(a)pyrene (BaP), has been routinely monitored in Western Europe since 2005 (the limit for its yearly concentrations is 1 ng/m^3). In Poland, in 2015, the highest yearly PM_{10}-bound BaP concentrations reached 15.6 ng/m^3 at some sites in southern and central Poland, and their average over 137 measuring sites scattered over the country (117 urban background, 7 suburban, 8 regional background, 3 traffic sites, and 2 sites affected by industrial emissions) was 5 ng/m^3 (GIOŚ, 2016). Practically, neither indoor concentrations of PM-bound PAH (including BaP) nor the effects of outdoor on indoor PM-bound PAH concentrations have been investigated in Poland so far.

The US EPA recognizes sixteen PM-bound PAH as priority pollutants, and seven of them as carcinogens (Wang et al., 2010). The goal of the present study is to investigate the concentrations of PM_1-bound phase of these 16 PAH in two university teaching rooms and their dependence on the outdoor PM_1-bound PAH concentrations. Teaching rooms are usually smaller than, for example, public libraries, museums, theaters, *etc.*, and more intensely used (many people, usually during the better part of a day). The PM samples were taken in two Polish cities, Warsaw in central Poland, and Gliwice in southern Poland, differing with the structure of the PM emissions. Probable sources of PAH and the level of the health hazard to humans from the mixture of PAH (*i.e.*, equivalents: carcinogenic, CEQ, mutagenic, MEQ, and TCDD-toxic, TEQ) were also determined.

1. Material and methods

The PM_1-samples were collected in April–June 2015 to preclude intense winter emissions of PM and PAH from domestic coal-fired stoves (especially in Gliwice) and from heating plants, coal-fired as well. In whole Poland, the physicochemical properties of PM in winter (heating season) are entirely different from those in the rest of the year (Rogula-Kozłowska et al., 2013, 2016), probably because the PM from heating, moving with air masses, reaches even sites very remote from big PM sources (Rogula-Kozłowska et al., 2014). Also, the high ambient temperatures, insolation, and ozone concentrations of a hot summer period (July–August), favorable to the intense PAH transformations, and to the evaporation of light PAH from PM and their staying in gaseous form in the air (Srogi, 2007; Ravindra et al., 2008; Dvorská et al., 2011) were avoided. The air temperature in the measuring period was between 6.9–19.7°C (av. 14.9°C) in Gliwice and 7.5–23.3°C (av. 13.5°C) in Warsaw.

The 24-hr samples of PM_1 were collected simultaneously in the teaching rooms and their outsides, and simultaneously in Warsaw and in Gliwice. Both the rooms are located on the 3rd floor (approximately 8 to 10 m above the ground level). Neither air conditioning nor air cleaning equipment was installed in the rooms. They were heated with central heating radiators, there were no indoor PAH sources like smoking, cooking, *etc.* in the rooms. The tight PVC windows were opened only for airing before class and in the evenings during cleaning. Each room had a floor area of about 100 m^2, usually there were 20–30 people in it during class. The PM samplers were located in the room corners, about 2 m from the nearest wall, 5 (Gliwice) and 8 m (Warsaw) away from the windows and 6 (Gliwice) and 10 m (Warsaw) from the door.

All the outdoor samples were taken about 5 m above the ground level. The outdoor samplers were located 50 (Gliwice) and 70 m (Warsaw) – in a straight line – from the indoor samplers. In Warsaw, the distance to the nearest residential buildings, where coal and biomass might be combusted for heating, was 850 m; it was 350 m in Gliwice. The nearest coal-fired power stations were about 4 km away in Warsaw and 2.5 km in Gliwice. In European urban areas, an important source of PAH is road traffic (Ravindra et al., 2008). Both outdoor samplers were located about 200 m from busy roads. The samples were taken from Monday through Friday, when classes or lectures were held in the rooms. Altogether, 30 pairs of 24-hr samples of PM_1 were taken in each Gliwice and Warsaw (120 in total).

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