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# Indoor–outdoor levels of size segregated particulate matter and mono/polycyclic aromatic hydrocarbons among urban areas using solid fuels for heating

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## HIGHLIGHTS

- Air quality was monitored in six naturally ventilated homes for fuel combustion markers.
- Indications of non-biomass fuel combustion were observed.
- I/O distributions of individual PAHs were affected by the indoor combustion sources.
- Penetration of outdoor combustion PAHs to indoor air equalled from 5 to 36%.

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## ABSTRACT

Emissions from the fuel combustion in the energy production are causes of concern due to associated health risks, but little information is available on the impact of residential fuel burning on indoor air quality, where most of the human exposure occurs. In this complex study, concentrations of size-segregated particulate matter (PM), monocyclic and polycyclic aromatic compounds (MAHs and PAHs) at indoor and outdoor sites in six urban homes in the city of Kaunas, Lithuania, were determined over winter and summer sampling campaigns, specifically targeting the impact of the local fuel burning to the indoor air quality. PM levels observed in Kaunas during winter measurement campaign were higher compared to those in many other European settlements utilizing biomass for energy production. The particle size distribution analysis revealed that the major part of the PM mass in winter period consisted of fine particles ($PM_{2.5}$). Both MAH and PAH levels were higher in winter. The indoor to outdoor ratios (I/O) of MAHs and PAHs revealed specific patterns depending on the presence of emissions sources indoors. Irrespectively of the season, I/O values were <1, suggesting that in case of the absence of an indoor pollution, the dominant source of organic compounds was from the outdoor environment. In homes with no PAH source inside, the I/O ratio equalled ranged from 0.05 to 0.36, suggesting the penetrated portion of outdoor combustion particles to the indoor air.



## 1. Introduction

The burning of the solid biomass for the heat energy production is one of the main sources of renewable energy. The usage of biomass in the global primary energy balance is estimated to grow to 4.8 Gtoe by 2050, but some estimates account for 24 Gtoe biomass consumption (Williams et al., 2012). However, the biomass combustion can be a major source of emissions of fine particulate matter (PM), black carbon (soot), dioxins, volatile organic compounds (VOCs), including monocyclic aromatic hydrocarbons (MAHs, sometimes referred to as BTEXs – benzene, toluene, ethylbenzene, and xylenes), and polycyclic aromatic hydrocarbons (PAHs), and cause negative short and long-term health effects.

The EPA's 2005 National-Scale Air Toxics Assessment showed that total black carbon emissions from residential wood combustion in developed countries are estimated at about 4% of the total global inventory (311 Gg) and 16% of total residential emissions worldwide. The 2005 $PM_{2.5}$ inventory shows that cord wood stoves contribute about 52%, fireplaces 16%, hydronic heaters 16%, indoor furnaces 11%

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**Table 1**
Characterization of sampling locations, examined residential buildings and meteorological conditions during the study.

| # | Type of location | Housing density in the area, house/km$^2$ | Type of ventilation | Type of heating, fuel type[b] | Type of windows[c] | Occupancy | Average outdoor temperature, °C | Average wind speed, m/s | Average relative humidity, % |
|---|---|---|---|---|---|---|---|---|---|
| 1 | Urban | 1000–1400 | Natural | S–B, NG | P, DP | Unoccupied | −13 | 0.87 | 87 |
| 2 | Urban | 1000–1400 | Natural[a] | S–B, NG | W, D | Regular living by 2 persons | −4 | 3.72 | 91 |
| | | | | | | | 16 | 1.22 | 72 |
| 3 | Sub-urban | 400–600 | Natural[a] | S–B, WP | W, T | Regular living by 6 persons | 0 | 2.18 | 92 |
| 4 | Sub-urban | <400 | Natural[a] | HP | W, DP | Regular living by 5 persons | −4 | 2.18 | 86 |
| 5 | Urban | 600–1000 | Natural[a] | S–B, W | P, T | Regular living by 6 persons | 1 | 3.24 | 76 |
| | | | | | | | 13 | 2.66 | 78 |
| 6 | Urban | >1400 | Natural[a] | S–B, NG | P, T | Regular living by 5 persons | −1 | 3.17 | 67 |

Examined buildings.
[a] Temporary operation of mechanical ventilation in kitchen and bathroom.
[b] Type of heating, fuel type: S–B – stove-boiler, HP – heat pump, W – fire wood, WP – wood pallets, NG – natural gas.
[c] Type of windows: P – plastic, W – wooden, D – double pane glass, T – triplex pane glass; DP – double pane glass packs.

and pellet stoves and chimneys (free-standing outdoor fireplaces) the remaining 5%. In addition to BC and $PM_{2.5}$, residential wood combustion accounts for 44% of polycyclic organic matter emissions and 62% of the 7-PAHs (EPA, 2005).

Small scale units have more detrimental effects because of much higher emission factors of incomplete pollutants, which are often several orders of magnitude higher than that in centralized system. Studies on the pollution from residential wood burning in Denmark revealed that it is responsible for about 80 percent of Danish PAH emissions, 70 percent of $PM_{2.5}$, 60 percent of black carbon, 50 percent of dioxin and contributes significantly to emissions of VOCs and CO. In comparison, all Danish power plants emit about 2 percent of the total $PM_{2.5}$ emissions but produce more than 60 percent of the energy (Press-Kristensen, 2013).

Recent air quality studies indicate a rising worldwide concern due to the significant contribution of biomass combustion to ambient air pollution (Glasius et al., 2008; Gustafson et al., 2008; Hellén et al., 2008) and underline the importance of using efficient wood combustion technologies to improve the air quality in residential areas (Bari et al., 2010, 2011; Gonçalves et al., 2012; Huttunen et al., 2012; Kaivosoja et al., 2012; Meyer, 2012; Piazzalunga et al., 2013).

While the above presented studies addressed quantitative estimate of the contribution from the residential heating to the ambient air quality, very few have aimed at the investigation of the contribution of the products of fuel combustion to the indoor air quality. The specific products of fuel combustion and their indicators as well as size selective aerosol sampling allow both the qualitative and quantitative estimation of the penetration of these pollutants to indoor environment.

The target of this study was to report indoor and outdoor concentrations of size segregated PM, PAHs, and MAHs during the heating season in urban settlements. Specific aims of the study were to determine outdoor and indoor ratios of PAHs in size-segregated particulate matter, diagnostic ratios of PAHs, as well as indoor/outdoor ratios of PM, PAHs and MAHs, implying the tentative exposure of the residents to the pollution from outdoors.

## 2. Methods

### 2.1. Sampling locations

The city of Kaunas (pop. 311,100; total area 157 km$^2$), Lithuania, is situated on the confluence of the Nemunas and the Neris rivers. The central part of the city has a well-developed network of a collective heating system. During recent years, due to rising costs of fossil fuels, a shift from the collective heating to the in-house energy production became more prominent. These individual energy production units mostly depend on a solid fuel (usually wood chips or pellets, sometimes hard coal), although natural gas supply lines are also widely available. A common phenomenon which occurs in the districts of lower income is the incineration of the calorific fraction of municipal waste, potentially contributing to serious emissions of aromatic species of hydrocarbons to the ambient air.

Daily samples of PM, PAHs, and MAHs were collected outdoors and indoors over the six-week period in January, February, and March of 2013. The identical analysis has been conducted in L2 and L5 in August–September, 2013. In each case, samples were collected over twenty-four hour periods during weekdays.

Six sampling locations (single-family homes) in Kaunas were chosen (Fig. S1). The selected locations represented different districts of Kaunas city and had a typical layout of houses. Four locations may be classified as "urban" and two locations as "suburban" sampling sites (see Table 1). The first and the second locations (henceforth referred to as L1 and L2) were "urban" sites situated at the city districts with high a density of one-family (mostly lower income) houses (1000–1400 houses/km$^2$) built in the period of 1930–1970. The majority of the houses had a poor thermal insulation and utilized natural gas or solid fuel stoves and boilers.

The third location (L3) was situated in the southern part of the Kaunas city. One-family houses were built during the period of 1960–1980; the average density of buildings was 400–600 houses/km$^2$. The vast majority of buildings have poor thermal insulation and are heated by wood or other solid fuels.

**Table 2**
$PM_{2.5}$ concentrations (μg/m$^3$) at sampling locations.

| | L1 W | | L2 W | | L2 S | | L3 W | | L4 W | | L5 W | | L5 S | | L6 W | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Indoor | Outdoor | Indoor | Outdoor | Indoor | Outdoor | Indoor | Outdoor | Indoor | Outdoor | Indoor | Outdoor | Indoor | Outdoor | Indoor | Outdoor |
| Median | 90.9 | 38.7 | 39.6 | 32.4 | 8.6 | 15.6 | 41.3 | 33.2 | 40.8 | 24.2 | 59.7 | 56.5 | 7.8 | 8.8 | 24.3 | 14.8 |
| Minimum | 41.3 | 25.5 | 11.1 | 3.3 | 6.6 | 12.4 | 37.2 | 29.5 | 26.1 | 18.9 | 51.2 | 2.4 | 6.0 | 7.1 | 3.2 | 7.4 |
| Maximum | 183.0 | 248.2 | 112.4 | 61.5 | 10.1 | 18.6 | 75.3 | 60.0 | 50.0 | 35.8 | 99.5 | 67.3 | 13.6 | 11.1 | 51.8 | 23.0 |

W – winter measurement campaign, S – summer measurement campaign.