



PAHs in Household Floor Dust Collected in Amman, Jordan

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Abstract

People are exposed to polycyclic aromatic hydrocarbons (PAHs) outdoors and indoors. Although there is a fair amount of literature review concerning exposure to settled dust, and in particular, to PAHs indoors, no attempts have been made to report contamination in household settled floor dust in Jordan. In this study we aimed at identifying the source origin of the PAHs as indoors or outdoors inside eight selected dwellings in Amman, Jordan. We also aimed at investigating the spatial variation of the analyzed PAHs concentrations within Amman. The floor dust samples were collected from the entrance and living room areas. The total PAHs concentrations at the living room area ranged from 65422 ng/g to about 641 ng/g. At the entrance area, the highest total PAHs concentration was about 9266 ng/g and the lowest was about 241 ng/g. The differences among dwellings for the total and individual PAHs concentrations confirms that each dwelling has its own environmental conditions and we cannot make a general conclusion with respect to the source origin as indoors or outdoors. The spatial variation of the PAHs concentrations showed that the north-eastern part of Amman had lower PAHs concentrations than that observed in the south-western part. This agrees well with a previous study about the spatial variation of fine aerosol particle number concentrations measured within the city during the same period by Hussein et al. who reported that fine particle number concentrations in the north eastern part of the city is lower than that in the south western part.

Keywords: Indoor contamination; Spatial variation; Source origin

Introduction

People spend more than 80% of their time indoors [1-6], where they are exposed to a variety of pollutants that are either originated indoors or simply penetrated across the build shell from outdoors. Indoor sources of contamination are mainly household chemicals, toys, electrical devices, building and construction materials, furnishing, cosmetics, biocides, combustion processes and emissions related to human activities such as smoking, cooking, etc. [3,6]. Outdoor sources of contamination may include vehicle exhaust emissions, industrial emissions, and natural aerosols (dust, bioaerosols, etc.). Eventually, contamination settles down onto indoor surfaces.

Polycyclic aromatic hydrocarbons (PAHs) are one of the most perilous pollutants that can be found in indoor floor dust. PAHs are organic compounds with two or more fused aromatic rings [2,6]. They are usually formed during smoking and incomplete combustion of coal, oil, and gas [7]. Although there are many different types of PAHs, the United States Environmental Protection Agency listed only seven PAHs in the Priority Chemical list: acenaphthene, acenaphthylene, anthracene, benzo [g,h,i] perylene, fluorene, phenanthrene, and pyrene. Most of PAHs are toxic, carcinogenic, mutagenic, and at the same time are persistent and bio accumulative [2,6,8-17]. For example, the Agency for Toxic Substances and Disease Registry announced that benzo[a] pyrene (B[a]P) and benzo(a)anthracene (B[a]A) are among the most predominant carcinogenic PAHs (e.g., Ref. [18]). Dibenzo[a,h]pyrene and dibenz [a]anthracene can be even 100 and 5 times respectively more carcinogenic than benzo[a]pyrene [19]. Miller et al. [20] and Perera et al. [17] made evidence that traffic-related-PAHs have a potential risk for asthma for children.

There are many reports and studies about PAHs in indoor floor dust collected worldwide [14,18,21-23]. However, and according to

our knowledge, there is nothing found about that in Jordan. Therefore, we prepared this study to investigate the PAHs content in household floor dust collected from dwellings within the capital region of Amman. We aimed at identifying the source origin of the PAHs as indoors or outdoors. We also aimed at investigating the spatial variation of the PAHs within the capital city Amman.

Materials and Methods

Dwellings and site locations

The measurement campaign took place in Amman, Jordan (Figure 1) during March 29-April 12, 2015. Floor dust samples were collected from 11 dwellings: five apartments, five detached houses, and a house. Two apartments and one detached house were excluded from the analysis because the amount of dust collected was not enough to pass through the chemical analysis.

Floor dust collection procedure

The floor dust samples were collected by using a regular household vacuum cleaner, where a small dust bag (nylon, 25 µm-155 mm × 73/38 mm, Allied Filter Fabrics Pvt. Ltd.) was mounted right after the

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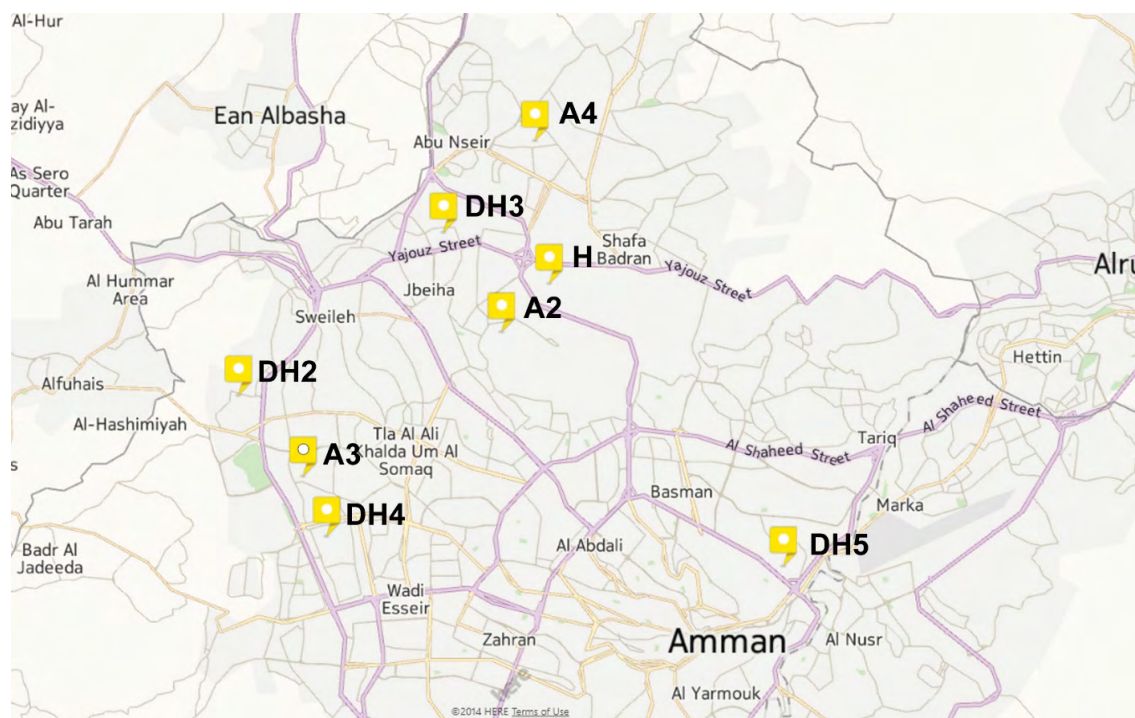


Figure 1: A map of Amman with site locations of the dwellings marked with yellow landmarks and the abbreviations of the dwellings as listed in Table 1.

Dwelling	Age [years]	Type	Level in Building	Sample Collection Location in Dwelling	Area [m ²], Type
DH2	10	Detached House	2 nd floor	Entrance	0.75 × 0.50, carpet
				Living room	1.00 × 1.00, carpet
A2	5	Apartment	2 nd floor	Entrance	0.91 × 0.60, carpet
				Living room	1.00 × 1.00, carpet
DH3	30	Detached House	Ground floor	Entrance (inside)	2.00 × 2.00, carpet
				Living room	2.00 × 2.00, carpet
DH4	15	Detached House	Ground floor	Entrance (inside)	0.60 × 0.60, carpet
				Living room	2.00 × 2.00, carpet
A3	4	Apartment	1 st floor	Entrance (inside)	1.00 × 0.50, carpet
				Living room	2.00 × 2.00, carpet
A4	3	Apartment	2 nd floor	Entrance (inside)	2.00 × 2.00, bare floor
				Living room	2.00 × 2.00, carpet
DH5	40	Detached House	Ground floor	Entrance (inside)	2.00 × 2.00, carpet
				Living room	2.00 × 2.00, carpet
H	10	House	Ground floor	Entrance	1.00 × 1.00, bare floor
				Living room	2.00 × 2.00, bare floor

Table 1: Features of the dwellings and floor, where the dust samples were collected.

main façade of the vacuum cleaner inside the tube. Two samples were taken from each house: (1) living room area and (2) main entrance. In order to collect enough dust amounts, the houses were not vacuum cleaned for 3-4 days before samples collection. The total area of floor dust collection was recorded; typically, we vacuumed 1 m² for each sample. Immediately after floor dust collection, the small dust bag was firmly closed and placed inside a nylon bag that was also zipped firmly. Prior to the GC-MS analysis, each dust sample was stored in a glass vial, wrapped with aluminum foil and stored in the freezer. In general, the amount of dust collected for each sample was more than enough for the chemical analysis.

A questionnaire was filled in by the occupants to provide detailed

information about their dwelling. The information in the questionnaire included: ventilation type, size of the kitchen, size of the living room, type of floor and carpets, cooking style and type, household appliances, heating system, and type of cleaning materials. It also included details about the occupants themselves (such as age) and smoking activities inside the dwelling. The type and age of each dwelling, the pavement type and area of the floor as well as the kind of the indoor location are given in the Table 1.

PAHs analysis

The dust samples were first manually and gently sieved to 500 µm size by using sieve 35 meshes (500 µm). Then they were prepared for the

chemical analysis by the GC-MS. The preparation included Pressurized Liquid Extraction (PLE) for 50-100 mg of sieved dust using a Dionex ASE350 under optimal conditions ($T=90^{\circ}\text{C}$, $P=1500$ psi, 5 minutes heating time, 4 minutes static time, and 50% flush volume); the number of static cycles was 3 times. In the clean-up process, the crude extracts were concentrated by using a Zymark Turbovap[®] II and purified by adding them to a florisil column (1 g); the column was washed with 8 mL of hexane to remove any lipids. PAHs were eluted with 15 mL 50% Hexane/Dichloromethane (Hex/DCM). The combined extracts were then turbo-vaped to 0.5-1.0 mL under a gentle stream of nitrogen and the PAHs were recovered by reconstitution in 100 μL of hexane (containing PAH recovery determination standard) prior to GC-EI/MS analysis.

In the GC-MS an Rxi- PAH capillary column (40 m \times 0.18 mm \times 0.07 μm) and helium as a carrier gas were used. 1 μL injection volume with pulsed splitless mode at a constant flow of 20 mL/min was employed. The injector temperature was set at 285°C and the oven temperature programme was as follows: 100°C (held for 1 min), first ramp at $37^{\circ}\text{C}/\text{min}$ to 210°C , second ramp at $3^{\circ}\text{C}/\text{min}$ to 260°C and third ramp at $11^{\circ}\text{C}/\text{min}$ to 320°C .

The mass spectrometer source was operated in the electron impact ionization mode (EI) at 70eV and 230°C whereas the mass analyzer was operated in the selected ion monitoring mode (SIM) at 150°C for better sensitivity, selectivity and matrix effect eliminating.

We focused on the identification of the following 13 priority PAHs: Phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benz[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[j]fluoranthene (BjF), benzo(a)pyrene [BaP], indeno[1,2,3-cd]pyrene (IcdP), dibenz[a,h]anthracene (DahA), benzo[g,h,i]perylene (BghiP).

We used anthracene-d10, benz(a) anthracene-d12 and perylene-d12 as internal standards. We also used phenanthrene-d10, pyrene-d10 and benzo(a)pyrene-d12 as surrogate standards. Concerning QA/QC check, SRM 2585 was running parallel to the samples to validate the analysis. Each sample was analyzed only once and there were no

duplicates. The recoveries were calculated as the recoveries of the surrogate standards and were in the range of 70% to 130%. Table S1 presents the detection limit (IDL) as well as the standard deviation (SD) of each PAH compound.

Results and Discussion

Total PAHs concentration

We considered the total PAHs concentrations in each dwelling for the living room and entrance areas separately (Figure 2 and Tables S2 and S3). This revealed the contamination level in the floor dust collected from each dwelling regardless to the type of the PAH.

Detached house 4 (DH4) had the highest total PAH concentration (65422.09 ng/g) at the living room area. The rest of dwellings had total PAH concentration lower than 10000 ng/g at the living room area (Figure 2 and Table S2). Basically, four dwellings (A2, A4, DH5, and H) had total PAH concentrations below 1000 ng/g (as low as 641 ng/g in dwelling A4) in the floor dust collected at the living room area (Table S2). Apartment 3 (A3) had the highest concentration of total PAHs (9266.27 ng/g) in the floor dust collected at the entrance area (Figure 2 and Table S3). Keeping in mind that the floor dust sample at DH4 and A3 collected at the entrance were inside the dwellings themselves, where the total PAHs collected at living room of these dwellings were the highest among all dwellings indicating that a significant fraction of the total PAHs' concentration at the entrance area was basically from indoor sources. The lowest concentration of total PAHs (241 ng/g) at the entrance area was observed in house (H), which seems to be the least contaminated from outdoor surrounding compared to its living room concentration of total PAHs (702.87 ng/g).

In the four dwellings (DH2, A2, A3, and DH5) the total PAHs concentration at the entrance area was higher than that at the living room area where the concentration ratio of total PAHs between living room to entrance area was 0.29, 0.64, 0.79, and 0.79 respectively. For the rest of the dwellings (DH3, DH4, A4, and H); the opposite occurred (Figure 2 and Table S3), where the total PAHs concentration ratio between living room to entrance area was 1.5, 9, and 2.9 for DH3, DH4,

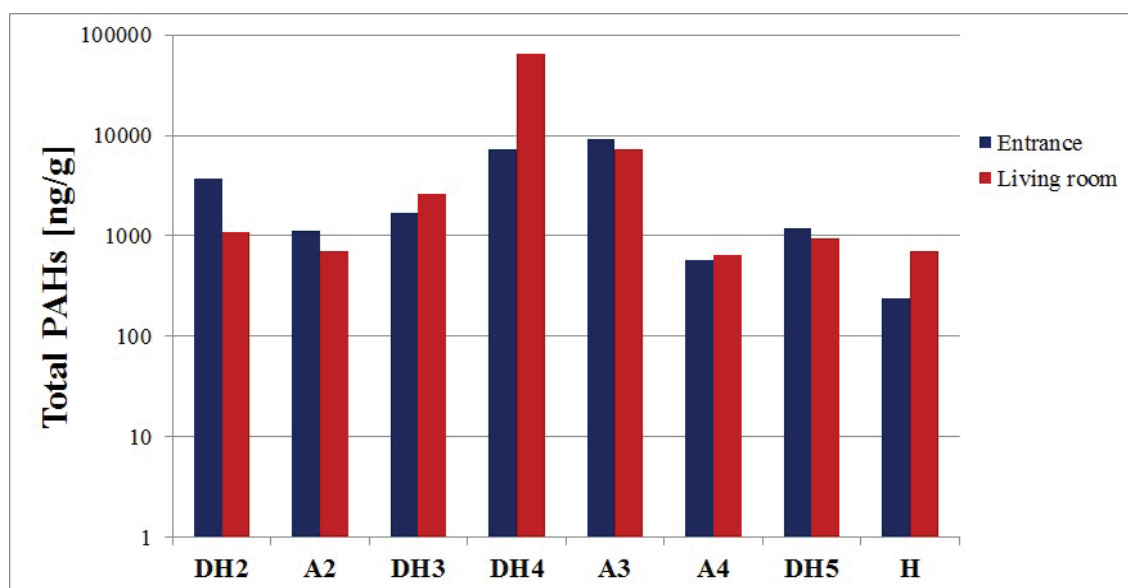


Figure 2: Total PAHs concentration in the living room and the entrance area for each dwelling.

and H; respectively. For A4, the ratio was about 1.1. Such difference between the entrance and living room total PAHs' concentrations in each dwelling provides a good indication about the PAHs source origin (as either indoor or outdoor). While this fact is obvious, still it needs to be interpreted carefully as will be discussed in the next section for the individual PAH concentrations for each dwelling.

According to Figure 1 and Tables S2 and S3, the total concentrations of PAHs in both living room and entrance in the north-eastern sites (A4, H, A2, and DH5; except for DH3) tended to have lower total PAHs concentrations than the south-western sites. This could be attributed to lower concentration of fine particles in this region [1] in addition to the fact that the major PAHs found indoor dust including Fluoranthene, phenanthrene, and anthracene have particle gas phase distribution [24].

PAHs concentrations

In general, DH4 exhibited the highest concentrations for all PAHs at the living room area followed by dwellings A3 and DH3. Among all PAHs concentrations, fluoranthene, pyrene, and phenanthrene were the highest at the living room area with values as high as 26630, 15780, and 14413 ng/g respectively in dwelling DH4 (Figure 3a and

Table S1). Fluoranthene and phenanthrene are associated with fossil fuel combustion. In addition, phenanthrene is also linked to tobacco smoke emissions [25]. In our case, high values of fluoranthene and phenanthrene can be related to the fact that DH4, A3, and DH3 were located nearby busy streets and their residents used to apply natural gas for cooking (stir-frying, deep frying, stewing, streaming, grilling, boiling, roasting, and/or baking); which was couple of hours a day [26-29]. As for the elevated concentration of fluoranthene in DH4 (almost 27000 ng/g), it could be the result of using a kerosene heater, occasionally smoking inside, and stir-frying/deep frying (3 hours/day). Benzo[a]pyrene, benzo[j]fluoranthene and chrysene were also the highest in DH4. The sources can be from residential charcoal uses, cigarette smoking, oil heating, and coke emissions (e.g., Ref. [6,30,31]).

Anthracene, benzo[a]anthracene, benzo[j]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, and dibenz[a,h]anthracene concentrations were below the detection limit in some dwellings (Figure 3a). Among all PAHs concentrations that were detected, the lowest concentrations observed at the living room area of dwelling A4 located in very quiet neighborhood were 11.5 ng/g and 9.6 ng/g for benzo[j]fluoranthene and benzo[a]pyrene, respectively. In fact, A4 was occupied by a nonsmoking resident, who rarely cooked at home

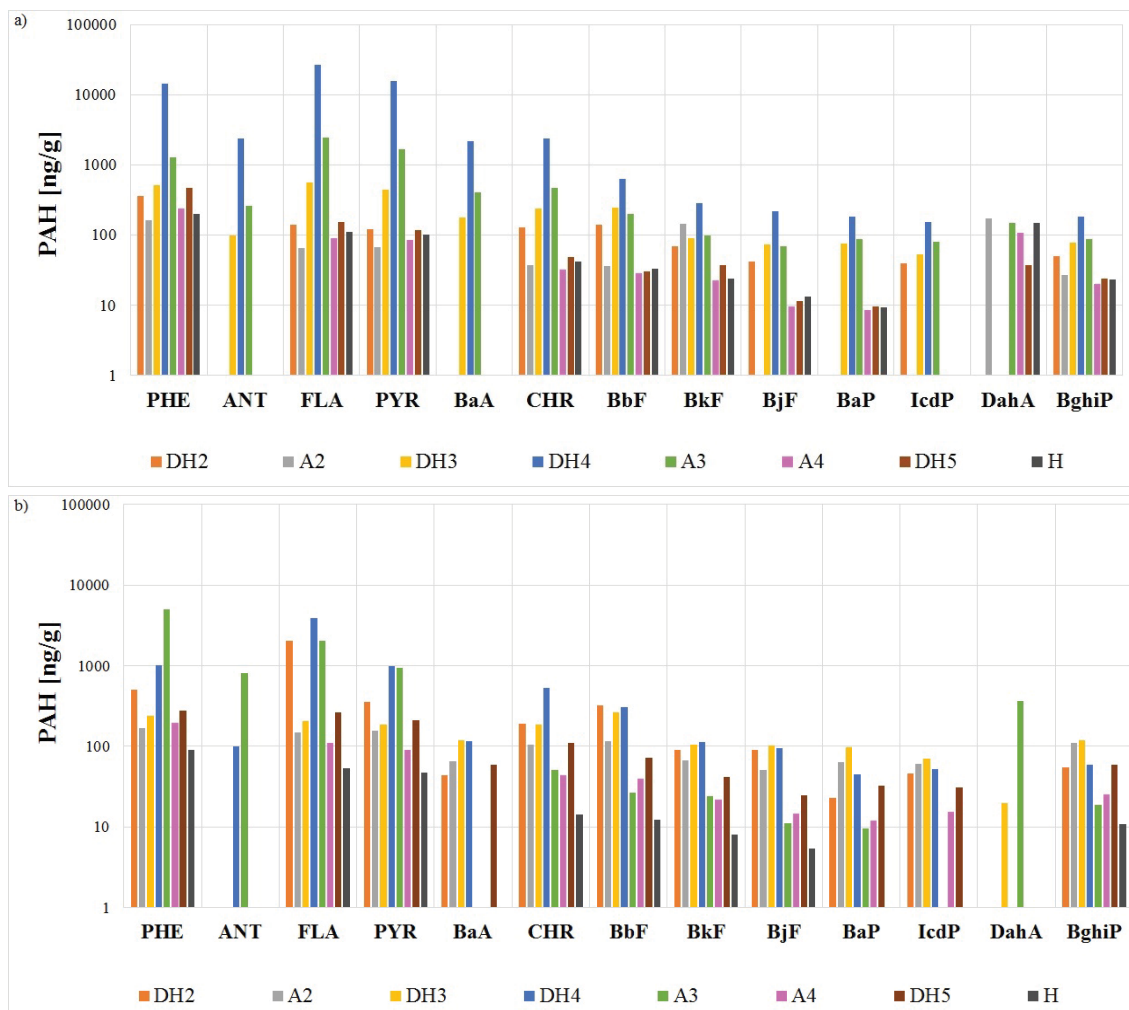


Figure 3: Individual PAH concentration in the floor dust collected at (a) living room area and (b) entrance area.

and occasionally used an electric heater as a replacement for the natural gas heater, which was rarely used.

The entrance area in dwelling DH4 exhibited the highest concentrations of fluoranthene (3879 ng/g) whereas dwelling A3 had the highest concentrations for phenanthrene (4973 ng/g) and anthracene (806 ng/g) as demonstrate in Figure 3b and Table S3. Both DH4 and A3 had the highest concentrations for pyrene; 997 ng/g and 946 ng/g, respectively. The dwelling H had the lowest PAHs concentrations. Dibenz[a,h]anthracene was below the detection limit for all dwellings except for DH3 and DH5. The entrance area in DH2, A2, and H was located outside the building whereas it was inside the building for the rest of dwellings. It is, therefore, obvious that the PAHs concentrations at the entrance area at those dwellings (i.e., DH2, A2, and H) are not affected by the indoor emissions; and thus, they will not have a direct relationship with the PAHs concentrations observed at the living room area. This factor will be considered in the next section for the living room-to-entrance PAHs concentration ratio interpretation and discussion.

Living room-to-entrance PAHs concentration ratios

The living room-to-entrance PAHs concentration ratio is an indication for the PAHs sources as from outdoor origin or from indoor origin [14,32]. For example, this ratio was larger than 1 for all PAHs observed in dwelling DH4 (Figure 4); indicating that the PAHs observed at the living room area are mainly from indoor origin. For dwelling A3 only phenanthrene, anthracene, and dibenz[a,h]anthracene had living room-to-entrance PAHs concentration ratios less than 1; indicating that those PAHs originated from outdoor sources. It is worth to mention here that for the above mentioned dwellings (i.e., DH4 and A3) the floor dust samples (both entrance and living room) were collected from inside the dwelling. This indeed might influence the PAHs concentration at the entrance area to be strongly affected by the indoor sources; however, a higher concentration at the living room area than those at the entrance for those dwellings is strong evidence that PAHs originated from an indoor source.

To get a better view about the living room-to-entrance relationship (i.e., indoor-to-outdoor ratio) for the observed PAHs; dwelling DH2 entrance floor dust samples were taken from the outside of the dwelling (Figure 4). For this dwelling, the living room-to-entrance

PAHs concentration ratios were less than one for all PAHs. Dwelling A2 also reflected the same view about the ratios except for benzo[k]fluoranthrene, concluding apparently that the PAHs observed in these two dwellings are from outdoor sources. Contrary to this was dwelling H where the living room-to-entrance PAHs concentration ratio was 2.9 indicating strongly that PAHs originated indoor even though entrance floor dust samples were taken from the outside of the dwelling.

Conclusion

We analyzed the PAHs concentrations in floor dust collected at the entrance and living areas in eight dwellings distributed within Amman, Jordan. We aimed at identifying the source origin of the PAHs as indoors or outdoors for each dwelling. We also aimed at investigating the spatial variation of the analyzed PAHs concentrations within the capital city Amman.

The total PAHs concentration in one of the houses was as high as 65422 ng/g at the living room area and it was lower than 10000 ng/g for the rest of dwellings; the lowest total PAHs concentration was about 641 ng/g. As for the entrance area, the highest total PAHs concentration was about 9266 ng/g and the lowest was about 241 ng/g. The total PAHs concentration at the entrance area was higher than that at the living room area for four dwellings. The differences among dwellings for the total and individual PAHs concentrations confirms that each dwelling had its own environmental conditions and we cannot make a general conclusion about those dwellings with respect to the source origin as indoors or outdoors. Therefore, a larger study to include more dwellings and more frequent sample collection is needed.

The spatial variation of the PAHs concentrations indicates that the north-eastern sites tended to have lower total PAHs concentrations at both the living room and entrance areas than those located at the south-western sites. This confirms the spatial variation of fine aerosol particle number concentrations measured within the city during the same period; Hussein et al. reported that fine particle number concentrations in the north eastern part of the city is lower than that in the south western part. It is very well know that fine particles in the urban atmosphere is associated with combustion sources (mainly traffic emissions), which are also a source of PAHs that settles down in floor dust.

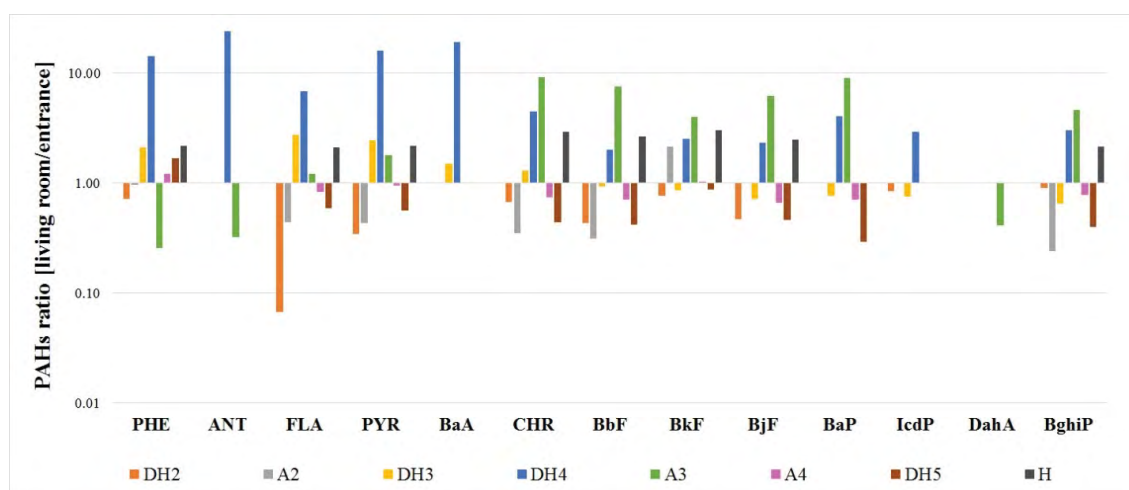


Figure 4: Individual PAHs concentration ratios between living room and entrance areas.

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