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Contamination and Potential Risk Assessment of Polycyclic Aromatic Hydrocarbons (PAHs) and Heavy Metals in House Settled Dust Collected from Residences of Young Children

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Abstract: House settled dust (HSD) contains various hazardous materials, including polycyclic aromatic hydrocarbons (PAHs) and metals. Exposure to toxicants contained in HSD is of paramount concern especially in the case of young children, due to their particular behavioral characteristics. In this context, extracts of sieved vacuum cleaner dust from 20 residences with young children were examined for the presence of PAHs and trace metals, in Athens, Greece. The results indicated that PAHs and metals were ubiquitous in the studied residences. The calculated enrichment factors (EF) of trace metals indicated that Cu, Se, Zn, Hg, Cd, and Pb were mainly of anthropogenic. According to the PCA analysis, the main sources of household dust were: smoking inside the houses, combustion processes, resuspension of soil dust, and vehicle traffic. In general, the cancer risk due to PAHs exposure was found lower than the threshold value. The ingestion of house dust was the most important route of exposure to metals. The dose of almost all elements for the children was found 1–2 orders of magnitude lower than the corresponding reference values. Both the carcinogenic and noncarcinogenic risks of exposure were within the safety limits.

Keywords: settled house dust; PAHs; metals; enrichment factor; sources; health risk assessment

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1. Introduction

Indoor air pollution plays a key role in human health since people spend the largest part of their time in indoor environments and 70–80% of their day in residential environments [1]. Indoor air and dust are the two main pathways ways of exposure to environmental contaminants. Depending on the nature of the contaminant itself, other routes like dietary exposure could also be of great importance. Compared to indoor air, indoor dust is more suitable for estimating human exposure to various chemical substances [2]. Polycyclic aromatic hydrocarbons (PAHs) and trace metals are both ubiquitous in house settled dust (HSD). Young children are more seriously affected by toxicants in HSD due to their particular behavioral characteristics, such as hand-to-mouth behavior, crawling, frequent mouth breathing, and sucking or chewing dirty toys [3–5]. Considering that children spend almost all of their day at home and their breathing zone is very close to the floor, where residential dust tends to accumulate, makes them more susceptible to environmental stressors [6].

Indoor sources of PAHs include smoking, cooking, gas-fired appliances, and the penetration of polluted outdoor air, since the incomplete combustion processes, traffic, industrial emissions, and heating with fossil fuels constitute some of the major outdoor sources of PAHs [2,7–10]. Several PAHs are known or suspected carcinogens, most prevalent among them being benzo[a]pyrene (B[a]P) and benz[a]anthracene (B[a]A) [7]. Whilst PAHs have not been directly associated with asthma and allergies so far, results from a number of studies suggest that traffic-derived PAHs and those emitted from smoking, cooking, and space heating may increase the risk for asthma-related symptoms and seroatopy in children [11].

Trace metals in indoor settled dust originated from both indoor and outdoor sources. External sources include soil, road dust, industrial and vehicular emissions, while decorative paints, consumer and cosmetic products, appliances, and combustion products of cooking, heating, and smoking constitute important indoor sources [12,13]. Inhalation, dust ingestion, and dermal contact are the main routes of human exposure to toxic metals, while it has been proved that incidental oral ingestion constitutes the most important exposure pathway, especially in the case of young children [12,14]. Taking into account that metals accumulated in the tissues and internal organs of the human body, affecting the central nervous system and acting as cofactors or promoters of other disorders, makes infants and young children more vulnerable to metal exposure when compared to adults [15,16]. In particular, trace metals such as Cd, Cr, Ni, and Pb can have cumulative effects, causing growth retardation in children, kidney disease, cancer, and several other adverse health effects [17].

To the best of our knowledge, there has been very limited research in Europe to examine the concentrations and sources of PAHs and trace metals in indoor dust [18–20] and even scarce focusing on residences with infants and young children [7]. Additionally, the present work is the first of its kind to be conducted in Greece. The objectives of this study were: (a) to identify the concentrations and profiles of PAHs and trace metals in indoor dust collected from Greek households, (b) to examine the relationship between the household characteristics and occupant's activities with PAH and trace metals concentrations in indoor dust, (c) to determine the sources of PAHs and trace metals in indoor dust of Greece, (d) to conduct a health risk assessment and to evaluate the cancer risks of exposure to PAHs and metals in residential environments.

2. Material and Methods

2.1. Study Design and Dust Sample Collection

In the present study, twenty residences located in various areas across the Athens region were investigated. Participants with a child below three years of age were eligible and were recruited both through advertising via flyers in several facilities of interest and word of mouth. The field campaigns took place during the warm season of 2015 (June–October). Before starting the sampling procedures, an informed consent form was signed by each participant. Vacuum cleaner bags were collected from all the 20 residences. The residents were asked to vacuum floors only but they were not limited to certain rooms. The idea behind that is that the residents carry the dust through movement from one room to another. The bags were removed from the vacuum cleaner, placed in zip-seal plastic bags, and transported to the laboratory where they were stored at −20 °C, as described by Zhu et al. [21]. The samples were then analyzed for 25 PAHs and 15 trace metals.

In addition, detailed data regarding the building characteristics, such as location and type of the residence, potential nearby pollution sources and presence of attachedgarage, were recorded. Participants were asked to fill out an additional questionnaire concerning the characteristics of the residences (e.g., age of the house, previous water damage, presence of pets, smoking status).

2.2. Samples Clean Up and Preparation

One day before the beginning of the analysis, the samples were placed in a fume hood to reach room temperature. Subsequently, each dust sample was sieved to achieve homogenization. At the end of the process, the sieved dust was weighed using a high precision electronic scale (GIBERTINI, E42S-B, Novate Milanese (MI), Italy) and stored at −20 °C until analysis.

2.3. Laboratory Analysis of PAHs

Out of the 20 dust samples that were collected, 3 were destroyed during the analytical process. Therefore, the results that were obtained from 17 residences are subsequently presented. The dust extracts were analyzed for 25 PAHs using gas chromatography/mass spectrometry (GC/MS), including naphthalene (Nap), 2-methylnaphthalene (2M-Nap), 1-methylnaphthalene (1M-Nap), 1,2-dimethylnaphthalene (1,2 DM-Nap), 2,6- dimethylnaphthalene (2,6 DM-Nap), acenaphthene (Ace), 2,3,5-trimethylnaphthalene (2,3,5-TM-Nap), acenaphthylene (Acy), anthracene (Ant), benzo[a]anthracene (B[a]A), benzo[a]pyrene (B[a]P), benzo[e]pyrene (B[e]P), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F),benzo[g,h,i]perylene (B[ghi]Per), chrysene (Chry), dibenzo[a,h]anthracene (dBaAnt), fluoranthene (Fla), fluorene (Fl), perylene (Per), indeno (1,2,3-c,d) pyrene (IndP), phenanthrene (Phe), dibenzothiophene (dbt) pyrene (Pyr), 3,6-dimethylphenanthrene (3,6-dMePhe), 1-methylphenanthrene (1-MePhe). The analysis of the polyaromatic hydrocarbons was performed according to ISO12884 with the use of the certified material NIST Urban Dust (1649b). The main steps of the analytical procedure are the following. Samples were extracted in a Soxhlet extractor for 24 h at a reflux rate of about 4 cycles per hour. Before the extraction, deuterared PAHs (d8-Nap, d10-A, d10-Phe, d10-Chr, d10-Pyr, d12-B[ghi]P and d12-Perylene) were added as internal standards to monitor recovery. Subsequently, the extracts were concentrated in a rotary evaporator, loaded onto activated silica gel column chromatography, and eluted with n-hexane and n-hexane/dichloromethane (3:2). PAHs fraction was concentrated under a gentle stream of nitrogen, and an aliquot was analyzed by GC/MS (Agilent Technologies 7890A GC). Twenty-five PAHs were detected, including a group of suspected carcinogens PAHs.

2.4. Laboratory Analysis of Trace Metals

All materials that came into contact with the samples were previously washed thoroughly, soaked in dilute HNO_3 (Merck, Darmstadt, Germany), and rinsed with ultrapure water of 18.2 MΩ cm (Millipore, Bedford, MA, USA). For the preparation of all required solutions, class A volumetric glassware was used. For the determination of As, Ba, Cd, Cr, Cu, Mn, Ni, Pb, Sr, V, the samples were digested with HNO_3 65% supra pure (Merck) with the subsequent addition of H_2O_2 30% (Merck), according to the procedure described by [22]. For Hg and Se, samples were acid digested with HNO_3 in Teflon vials closed and left overnight at room temperature. The following day, the vials were thermostated at 80 °C for 3 h. The samples were allowed to cool at room temperature before their dilution with Milli-Q water. The digested samples were analyzed through inductively coupled plasma mass spectrometry (ICP-MS) by a Thermo Scientific ICAP Qc (Waltham, MA, USA). Measurements were carried out in a single collision cell mode, with kinetic energy discrimination (KED) using pure He. Matrix induced signal suppressions and instrumental drift were corrected by internal standardization (^{45}Sc, ^{103}Rh). Analyses of Al, Fe, and Zn were carried out by flame atomic absorption spectrometry (FAAS) (SpectrAA 200; Varian, Mulgrave, Vic, Australia), following digestion of the samples with a mixture of HNO_3 (65%), HCl (30%) and HF (40%). The calibration curves matched the matrix (acidity) of the samples.

2.5. QA and QC

2.5.1. PAHs

The quantification of PAHs was carried out through a linear calibration curve, which was built on matrix-matched standards (dust on filters). For the determination of the curve a standard polyaromatic solution containing secondary PAHs derivatives was used as internal standards. The solutions were of known concentration (0.05–10 ng/µL). The expanded uncertainty (Uexp k = 2) ranged from 6.3% to 23%, while the LOD of the method ranged from 0.6 pg/µL to 12.8 pg/µL.

2.5.2. Trace Metals

The method detection limits (MDLs) were within the range 0.500–5.00 ng g^{-1} for ICP-MS determined elements and 25.0–50.0 µg g^{-1} for FAAS determined elements. For statistical calculations, values below the MDLs were assigned the method detection limits divided by $\sqrt{2}$. The quality assurance was provided by analyzing the certified reference material (CRM) NIST 1649a (urban dust), which includes reference values for selected trace elements.The recoveries for As, Ba, Cr, Cu, Fe, Mn, Ni, Pb, V, and Zn in the CRM were in the range ±20%. Further, recovery efficiency for spiking sample analysis was ±25% for all elements.

2.6. Statistical Analysis

Statistical analysis was performed with SPSS 22 software (SPSS Inc., Chicago, IL, USA) for Microsoft Windows®. The nonparametric Kruskal–Wallis test was used to examine whether there were significant differences in chemical concentrations among residences with different household characteristics and different occupant's activities. Statistical significance was set at two stages $p < 0.05$ (95% confidence interval) and $p < 0.1$ (90% confidence interval). Pearson's correlation coefficient analysis and principal component analysis (PCA) identified the relationship between contaminants and possible sources. PCA constitutes one of the most common multivariate statistical methods widely used in dust contamination studies [2,9,17,23].

2.7. Enrichment Factors

In order for the metal enrichment in house dust and possible natural or anthropogenic sources to be determined, enrichment factors (EF) were calculated using the Equation (1).

$$EF = \frac{\left(C_x/C_{ref}\right)_{sample}}{\left(C_x/C_{ref}\right)_{background}} \quad (1)$$

where C_x/C_{ref}: the ratio of concentrations of trace metal to the corresponding concentration of a reference metal in the sample and background. In the present study, Al was selected as reference metal, as it is considered that anthropogenic activity contributes the least to its presence. In general, as long as the enrichment factor is less than 10, the natural origin of the elements dominates, while for values greater than 10, their enrichment due to anthropogenic activity is important due to anthropogenic activity [15,23], especially for metals for which the factor is estimated to be greater than 40.

2.8. Cancer Risk Assessment of PAHs in Settled House Dust

The estimation of the excess lifetime cancer risks for young children associated with nondietary ingestion of PAHs in settled house dust was conducted, according to the methodology of Maerterns et al. [18], using the following equation:

$$\text{Lifetime cancer risk} = \sum_{i=1}^{n}\left(\frac{(C_i \times PEF_i \times IR \times EF \times SF \times AF}{BW \times 1000}\right) \quad (2)$$

where C: the concentration (µg/g) of each of the carcinogenic PAHs (B[a]A, B[a]P, B[b]F, B[k]F, Chr, D[ah]A and Ind), PEF (Potency Equivalency Factor): the factor that expresses the potency of each PAH in terms of B[a]P which were as follows: B[a]A = 0.1, B[b]F = 0.1, B[k]F = 0.1, Chr = 0.001, D[ah]A = 5 and Ind = 0.1, IR (Ingestion Rate): Daily ingestion rate of dust which was considered as 0.05 g/day and 0.1 g/day, EF (Exposure Factor): the rate of exposure, which was calculated taking into account that a preschool-aged children is active for 12 h (while the remaining 12 asleep) and assuming that is exposed to these concentrations until the age of 5, BW (Body Weight): the average body weight, which was considered equal to 13 Kg, SF (Slope Factor): the estimate of the probability of a response occurring per unit intake of the PAH over a lifetime and for the present study an oral slope factor for B[a]P equal to 7.3 (mg·Kg/day) was used, AF (Adjustment Factor): the factor accounts for children's exposure during early life stages. For exposure to carcinogens, U.S.EPA recommends: AF = 0 for children up to 2 years of age and AF = 3 for children between 2 and 15 years of age. It has been estimated that preschool-aged children (1–5 years old) ingest 0.05 to 0.1 g of dust per day, depending on the season and the amount of time spent indoors [18]. For this reason, two different scenarios for the calculation of the risk of carcinogenesis were included: a moderate exposure scenario (0.05 g/day) and a high exposure scenario (0.1 g/day). For the calculations, the average value of B2PAHs that was obtained taking into account the concentrations of all the studied residences, was used.

2.9. Health Risk Assessment of Metals in Settled House Dust

The present study aims to calculate the exposure of children and their parents to metals in house dust based on the assessment method of human exposure risk developed by US Environmental Protection Agency [24]. Residents are exposed to dust through three main pathways: ingestion, inhalation, and dermal contact. The dose received through each of the pathways was calculated by the following equations:

$$D_{ingestion} = \mathrm{C} \times \frac{\mathrm{IngR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \times 10^{-6} \tag{3}$$

$$D_{dermal} = \mathrm{C} \times \frac{\mathrm{SA} \times \mathrm{SL} \times \mathrm{ABS} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \times 10^{-6} \tag{4}$$

$$D_{inhalation} = \mathrm{C} \times \frac{\mathrm{InhR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{PEF} \times \mathrm{BW} \times \mathrm{AT}} \tag{5}$$

The available studies on the carcinogenic risk parameters in the current assessment standards have been conducted, taking into account only the pathway of inhalation [25]. Therefore, the daily average exposure for life through inhalation of a carcinogenic metal ($LD_{inhalation}$) was calculated by:

$$LD_{inhalation} = \frac{\mathrm{C} \times \mathrm{EF}}{\mathrm{PEF} \times \mathrm{AT}} \times \left(\frac{\mathrm{InhR_{child}} \times \mathrm{ED_{child}}}{\mathrm{BW_{child}}} + \frac{\mathrm{InhR_{adult}} \times \mathrm{ED_{adult}}}{\mathrm{BW_{adult}}} \right) \tag{6}$$

where C is the concentration (mg/kg) of the metal in the dust. IngR is the ingestion rate, estimated in the present study to be equal to 200 (mg/day) for children and 100 (mg/day) for their parents [26]. InhR is the inhalation rate and was considered as 5.71 for children and 19.02 for adults (m^3/day) [27]. Exposure frequency (EF), taking into account holidays, was assumed as 335 days per year. Exposure duration (ED) was taken as 5 years for children and 25 years for adults. AT is the averaging time (ED × 365 days for noncarcinogens and 70 × 365 days for carcinogens). BW is the average body weight (13 Kg for children and 70 Kg for adults). The surface area of skin exposure (SA) was considered to be 1150 cm^2 for children and 2145 cm^2 for their parents. The skin adhesive capacity (SL) was taken as 0.2 for children and 0.07 for adults. The skin absorption factor (ABS) was considered as 0.001 [15]. PEF is the particulate emission factor, in this study, 1.36×10^9 m^3/kg.

The doses that were calculated for each element and exposure pathway were subsequently divided by the corresponding reference dose (RfD) (mg/kg × day) to obtain the noncancer risk (HQ), while the dose was multiplied by the corresponding slope factor (SF) to calculate the level of cancer risk [15]. The hazard index (HI) is the sum of HQ. It is supported that if HQ or HI < 1, the risk is small, whereas in case HQ or HI >1, there is a noncarcinogenic risk. Risk refers to the risk of cancer, indicating the probability of cancer, which is usually expressed as the proportion of cancer population in unit population. The acceptable risk is within the range of 10^{-6} to 10^{-4} [15,27].

3. Results and Discussion

3.1. PAH and Trace Metals Concentrations

3.1.1. PAHs

Overall, the 25 PAHs were detected in all dust samples, except for perylene, which was not detected in six residences. The concentrations of all the individual PAHs are presented in Table 1. The total PAH concentration in indoor dust ranged between 1.4 and 7.3 μg/g, across the houses, with a median value of 2.2 μg/g. The house with the lowest total concentration was a recently renovated apartment, and its occupants had moved in about a month before the start of the campaign. This observation is consistent with previously conducted studies, where lower ΣPAH concentrations in newly constructed or renovated houses have been reported [18,21]. The house with the highest ΣPAH concentration was 10 years old, and the only distinct feature it had when compared to the other houses of the study was the presence of a large carpet in the living room. This finding is in agreement with the observations of Maertens et al. [18], who reported elevated total PAH concentration in a residence that was 90% carpeted. In general, the ΣPAH concentrations in the present study were lower with respect to previous studies. Specifically, the mean levels of total PAHs in indoor dust in two studies that were conducted in China were much higher as follows: 21.9–329.6 and 30.9 μg/g, respectively [2,9]. Similarly, higher concentrations were reported in USA (29.2 μg/g) [28,29], Canada (12.9 μg/g) [18] and Italy (5.11) [19].

Among the measured PAHs, phenanthrene was the most predominant, followed by fluorene, while perylene was the congener detected in the lowest concentrations in all the studied households. In terms of their molecular weight, PAHs are categorized in three main categories: PAHs with High Molecular Weight (HMW), Medium Molecular Weight (MMW) and Low Molecular Weight (LMW). HMW PAHs are in general more toxic and harmful with respect to the LMW, which are less toxic. In the present study, LMW PAHs were the predominant PAHs in indoor dust, accounting for 87.5 % of the total PAHs.

Table 1. Concentrations of polycyclic aromatic hydrocarbons (PAHs) in indoor dust samples (ng/g).

PAH (ng/g)	Mean	Median	SD	Min	Max
Nap	339	116	609	75.2	2479
2M-Nap	242	172	198	118	965
1M-Nap	126	91.6	107	61.1	518
Acy	15.7	13.2	7.82	8.24	39.4
1,2 DM-Nap	49.4	40.1	30.7	29.5	158
2,6 DM-Nap	11.7	9.08	7.04	6.43	33.2
Ace	94.6	78.0	69.0	60.8	353
2,3,5-TM-Nap	10.5	8.56	7.33	3.12	28.8
Fl	421	424	207	221	1116
DBT	25.4	21.2	20.9	4.96	91.8
Phe	925	905	465	528	2525
1M-Phe	67.9	45.8	79.0	14.9	358
3,6 DM-Phe	42.3	21.5	75.7	7.59	334
Ant	22.6	19.4	12.7	10.3	57.3
Flu	99.0	86.0	61.8	30.2	308
Pyr	100	74.4	117	20.1	542
BaA	11.3	8.25	12.2	1.68	54.9
Chr	45.3	27.2	72.9	4.25	325
BbF	20.7	16.9	19.9	2.01	90.4
BkF	10.7	7.22	12.3	1.01	55.7
Bep	14.0	9.79	14.8	1.49	65.9
Bap	7.06	5.45	5.69	1.05	24.0
Per	1.02	1.08	1.06	0.00	3.39
IndP	11.1	8.39	10.8	1.05	49.2
dBaAnt	2.06	1.69	1.83	0.39	8.39
B[ghi]Per	13.8	10.3	11.2	1.36	49.3

Out of the 25 measured PAHs, 16 (naphthalene, acenapthylene, acenaphtene, fluorene, phenanthrene, anthracene, fluoranthrene, pyrene, bez(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-c,d)pyrene, dibenzo(a,h)anthracene) have been designated high priority pollutants by Environmental Protection Agency (EPA), while 7 (bez(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-c,d)pyrene, dibenzo(a,h)anthracene) have been classified as human carcinogens by the U.S.EPA (2003), referred as B2 PAHs. The contribution of B2 PAHs was estimated equal to 5% of the total PAHs. Especially benzo (a) pyrene has been classified as particularly dangerous and has been extensively studied worldwide [7,18,30–32]. The German Federal Environmental Agency's Commission for indoor air quality has established the limit of 10 μg/g for exposure to B[a]P in house dust [18]. B[a]P concentrations in the present study were found 3 orders of magnitude lower than the above limit. Compared to the results of earlier studies, the concentrations of B[a]P found in this study were either lower [18,31,32] or comparable [7].

3.1.2. Trace Metals

The concentrations of the heavy metals identified in the dust samples are summarized in Table 2. Among the elements that were examined, Fe was found to have the highest concentration (4.9 μg/g), followed by Al (4.2 μg/g), Zn (0.4 μg/g), Cu (0.34 μg/g) and Ba (0.25 μg/g). The elevated concentrations of both soil-related (Fe, Al) and anthropogenic (Zn, Cu, Ba) elements in the dust samples implies that the dust accumulation in the residential indoor Environment is mainly affected by the resuspension of natural dust (soil) and road dust. The elements Hg and Cd had the lowest concentrations.

Table 2. Metal concentrations in indoor dust samples (μg/g).

Elements	Mean	SD	Min	Max
Zn	401	211	136	1031
Fe	4913	5650	851	26100
Al	4217	9391	269	44900
Hg	0.4	0.5	0.0	2.1
V	9.0	5.4	2.4	21.3
Cr	65.2	36.4	14.2	147
Mn	128	67.9	28.9	272
Ni	29.9	23.9	5.2	103
Cu	339	711	11.8	3051
As	4.0	4.4	1.3	19.8
Se	1.0	0.6	0.2	2.8
Sr	118	97.6	37.7	396
Cd	0.5	0.6	0.1	2.3
Ba	251	237.0	35.0	1130
Pb	46.1	71.5	6.5	343

The highest concentration of Fe (26.1 mg/g) was observed in a residence located in an urban area on a high circulation road. Indeed, Fe has been linked with traffic as it originates either from road dust resuspension or emissions from brake wear [33]. The highest concentration of Al (45 mg/g) was observed in a residence which was located in a rural area, while the highest levels of Zn, Cu, and Sr were detected in a ground floor house located in the Athens city centre, and thus significantly affected by vehicular emissions [34]. Due to the absence of established legislation, limits for trace metals in indoor dust were compared with concentrations reported in the literature. Compared to the metal concentrations reported in a recent study examining residential dust samples in Toronto, Canada, the levels of the present study were all higher (Cr (65.2 vs. 42 μg/g), Cu (339 vs. 136 μg/g), Ni (29.9 vs. 23 μg/g), Pb (46.1 vs. 36 μg/g), and Zn (401 vs. 386 μg/g), except for Cd (0.5 vs. 1.7 μg/g) [12]. Higher concentrations compared to those of the present work were also presented in an earlier study conducted in Japan [31], where Shakya 2013, reported lower concentrations for Cr, Ni, Pb, and Zn, but higher for Cd (0.5 vs. 8.2 μg/g) in Kathmandu, Nepal [32].

3.2. Relationship between PAH and Trace Metals Concentrations and Household Characteristics

The relationship between the individual PAH and trace metal concentrations and a series of household characteristics was examined.

3.2.1. PAHs

Compared to residences located in suburban areas, houses in urban areas were found to exhibit significantly higher concentrations of Anthracene (26.2 vs. 16.1 ng/g, $p < 0.05$) and other PAHs derived from combustion processes (Flut, Pyr, B[a]A, Chr, B[b]F, B[k]F, B[a]P, Ind και B[ghi]P), referred to hereafter as COMPAHs, such as B[a]P (8.5 vs. 5.8 ng/g). This is in agreement with the literature, as it has been reported that proximity to combustion sources such as vehicle exhaust emissions is probably the most important factor affecting the accumulation of PAHs in indoor dust [2,7,35]. The age of the house was found to affect both the Pyr (75 vs. 114 ng/g, $p < 0.05$) and all the B2PAH (87 vs. 140 ng/g, $p < 0.05$) concentrations, with the houses that have been built in the last 20 years presenting lower concentrations than the older ones. Houses on the ground floor presented higher concentrations of Flut, B[a]A, Chr, B[b]F, Ind, D[ah]A ($p < 0.05$), which are known to originate from combustion processes.

Both the B2PAH and COMPAH concentrations were considerably higher in the houses with an attached garage. Especially for BaP a significant difference was noticed (13.6 vs. 6.2 ng/g, $p < 0.05$). The existence of a metro or tram station nearby (< 200 m) was associated with increased concentrations of COMPAHs (479 vs. 217 ng/g, $p < 0.05$), especially those of B[a]A, B[a]P, Ind and D[ah]A. This is probably due to the heavy traffic and the parking stations that are located close to such stations. The presence of a carpet in the house was associated with increased concentrations of Nap (1319 vs. 209 ng/g, $p < 0.05$), while the habit of residents wearing shoes inside their home was associated with higher concentrations of COMPAHs (475 vs. 221 ng/g, $p < 0.05$), as well as B2PAHs (191 vs. 73 ng/g), especially those of B[a]P, Pyr, Chr, D[ah]A and B[ghi].

3.2.2. Metals

The location of the house seems to be an important factor in the presence of Zn ($p = 0.083$), Fe ($p = 0.071$), and Sr $p = 0.071$) in house dust, with houses located in urban areas having higher concentrations. Figure 1 illustrates the elevated concentrations of the aforementioned elements detected in urban areas, which are lower in suburban areas and especially in rural areas. This observation is explained by the common emission sources of these elements, which are traffic emissions and road dust resuspension. Also, houses located close to a highway (<200 m) indicated higher concentrations of Se (1.3 vs. 0.6 μg/g, $p = 0.073$).

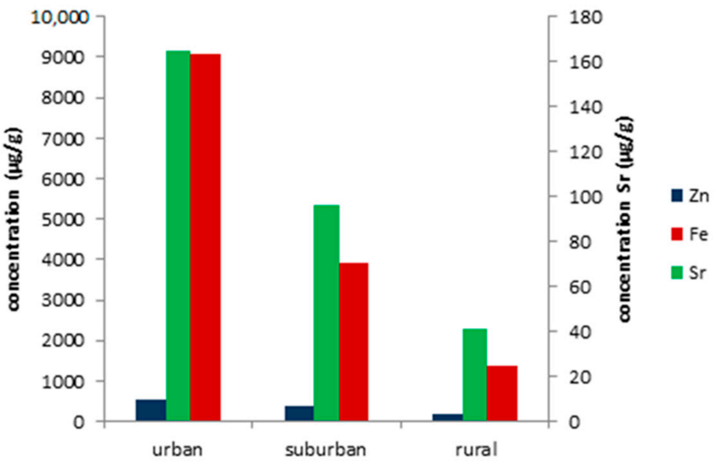

Figure 1. Concentrations of Fe, Zn and Sr in urban, suburban and rural areas.

Residences located either on the ground floor or the first floor indicated higher Hg concentrations with respect to those at higher floors (0.62 vs. 0.16 μg/g, $p < 0.05$). The age of the house was found to affect the levels of Sr, as higher concentrations in homes older than 20 years (128 vs. 77 μg/g, $p < 0.05$) were observed. In houses with a total area of less than 100 m^2, higher concentrations of Fe (6928 vs. 2526 μg/g, $p < 0.05$), Al (2691 vs. 1562 μg/g, $p < 0.05$), V (10.6 vs. 5.7 μg/g, $p < 0.05$) and Ba (327 vs. 133 μg/g, $p < 0.05$) were found. This fact can be related to the lower number of openings (windows and doors) to houses of smaller total area, which consequently leads to lower infiltration rates. Houses that used fireplaces as the main source of heating indicated significantly higher concentrations of Pb (343 vs. 32 μg/g, $p < 0.05$), probably due to the burning of painted or treated wood [33,34], while those that were heated using electricity were associated with significantly higher concentrations of Fe (10729 vs. 3529, $p < 0.05$) and V (12.8 vs. 7.3 μg/g, $p = 0.059$), likely due to dust resuspension occurring during the operation of air conditioning. In houses that remained closed (without the presence of occupants) for several hours during the day, higher values of As (8.7 vs. 2.5 μg/g, $p < 0.05$) and Sr (152 vs. 93, $p < 0.05$) were detected in household dust. The type of glazing appeared to affect Pb levels in household dust, as higher concentrations were observed in homes with single glazing compared to those with double glazing (57.8 vs. 46.2, $p = 0.078$). In terms of glazing on indoor Pb concentrations, we believe that the effect is not directly related to the type of glazing (or to be more accurate not only related to that) but on the type of the windows used. Double glazed windows are of newer technology and have been found in previous studies to significantly reduce the infiltration rates [35]. Since Pb is emitted from outdoor anthropogenic activities and resides in the fine fraction, it is expected that its indoor concentrations will be attributed mainly to infiltration. Finally, as with PAHs, the habit of residents wearing shoes inside their dwelling significantly affected the levels of Al (2684 vs. 1850 μg/g, $p = 0.076$) and Ba (275 vs. 217 μg/g, $p < 0.05$) in house dust. As both elements can be found in urban dust, there are transferred inside the residents by the dust that is found on their shoes.

3.3. Relationship between PAH and Trace Metals Concentrations and Occupant Activities

Apart from the household characteristics, the impact of the occupants' activities on the detection of PAHs and trace metals in dust was also examined.

3.3.1. PAHs

The analysis demonstrated that smoking inside the house significantly contributes to the increase of the concentrations of Phe (1275 versus 780 ng/g, $p = 0.0730.1$) and Ant (32.1 versus 18.7, $p = 0.073$), whereas no statistically significant differences for the other COMPAHs were observed. However, the concentrations of all COMPAHs in smoker's residences were found to be significantly higher than in nonsmoker's dwellings (Figure 2). In particular, Chr indicated 2.5 times higher concentration in smokers' homes (72 vs. 30 ng/g), while Pyr (153 vs. 63 ng/g), B[b]F (30 vs. 14 ng/g), B [k] F (16 vs. 7 ng/g) and B [a] *p* (10 vs. 5 ng/g) were found twice as high in smokers' houses as in nonsmokers. In a review that was conducted by Maertens et al. [36], a moderate but statistically significant relationship between smoking and PAHs concentration has been reported, while Langer et al. (2010) pointed out in their study that smoking was not a strong determinant in the presence of PAHs in house dust. However, other studies indicated no significant difference between smoking and nonsmoking dust [29,37], with Qi et al. [2] underlying that the effect of smoking on PAH concentration in dust remains uncertain. The use of air conditioning was strongly associated with both COMPAHs (548 vs. 227 ng/g, $p < 0.05$) and B2 PAHs (226 vs. 66 ng/g, $p < 0.05$). Finally, an inverse association between the concentration of ΣPAHs and cleaning activities was found. In particular, houses that were mopped more than once per week indicated significantly lower levels of ΣPAHs (3.4 versus 2.3 μg/g, $p < 0.05$). This finding suggests that frequent cleaning activities can help reduce the concentration of PAHs in household dust. This observation is in agreement with Maertens et al. [18], who reported a weak but statistically significant negative correlation between the frequency of vacuum use and the concentration of PAHs.

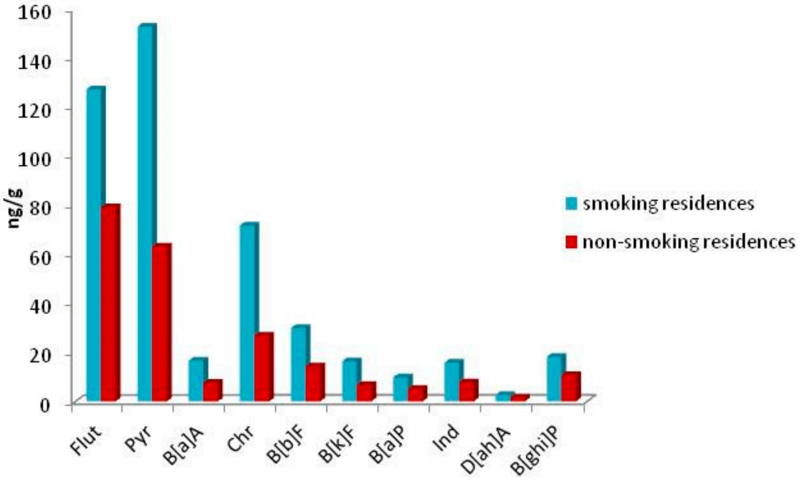

Figure 2. Concentrations of COMPAHs in smoking and nonsmoking houses.

3.3.2. Metals

In terms of residents' activities, smoking indoors was associated with increased concentrations of Cu (593 vs. 216 µg/g, $p < 0.05$), Sr (141 vs. 89 µg/g, $p < 0.05$) and Zn (422 vs. 379 µg/g, $p = 0.058.1$). Frequent cleaning activities (> 5 activities/week) were associated with increased Al concentrations (2801 vs. 1918 µg/g, $p < 0.05$) and the use of cleaning sprays was associated with increased levels of Pb (75.3 vs. 19.5 µg/g, $p < 0.05$). The use of air conditioning appeared to increase the concentrations of Cu (765 vs. 120 µg/g, $p = 0.070.1$), Zn (540 vs. 321 µg/g, $p = 0.092$) and Cd (1.0 vs. 0.3 µg/g, $p = 0.0090.1$). Finally, in terms of different cooking methods, frying was associated with increased levels of Ni (34.0 vs. 25.3 µg/g, $p < 0.05$) and Mn (147 vs. 100 µg/g, $p = 0.076$) in house dust, while frequent boiling (using of pot) was linked with higher levels of As (6.1 vs. 3.2 3 µg/g, $p < 0.05$) and Cd (1.1 vs. 0.3 3 µg/g, $p < 0.05$).

3.4. Enrichment Factors and Principal Component Analysis

Figure 3 presents EF of elements in the house dust samples. Several metals such as Fe, V, Mn, Sr, and Ba were of natural origin (EF < 10), while other metals such as Cu, Se, Zn, Hg, Cd, Pb (EF > 40) and Cr, Ni, As (20 < EF < 40) appeared to derive from anthropogenic processes.

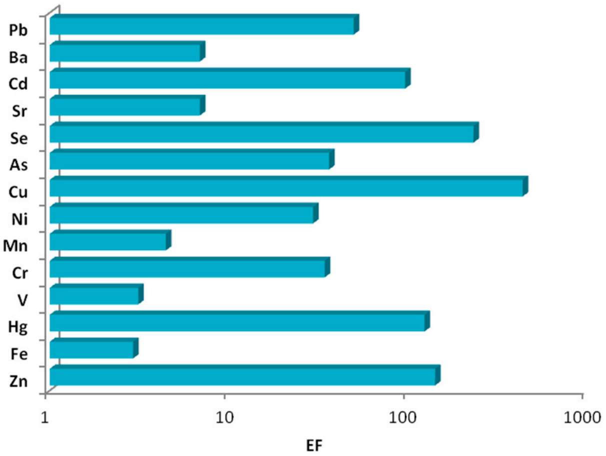

Figure 3. Enrichment factor (EF) of elements in house dust.

Principal Component Analysis (PCA) was performed for the PAHs and the metal contents of all dust samples. Eight PCs (Principal Components) were extracted with eigenvalues >1, by which 91.9% of the total variance was explained. The predominant components of each factor (>0.5) are presented in bold (Table 3).

Table 3. Results of Principal Component Analysis (PCA).

	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Factor 7	Factor 8
	Smoking	**Combustion Processes**	**Soil Dust**	**Vehicle Emissions**	**Road Dust**	**Abrasion of Plated Surfaces**	**Soil**	**Vehicle Emissions**
Eigenvalue	11.522	**4.840**	3.540	2.127	2.051	1.269	1.160	1.056
Cumulative %	38.41	**54.54**	66.34	73.43	80.27	84.50	88.36	91.88
Acy	0.187	**0.951**	−0.005	−0.035	0.068	−0.058	−0.108	0.132
Ace	−0.104	**0.962**	0.093	−0.025	−0.120	0.028	0.037	0.052
Fl	0.088	**0.973**	−0.026	−0.063	−0.030	−0.060	−0.087	−0.065
PH	0.068	**0.987**	−0.018	−0.060	0.027	−0.027	−0.049	−0.057
Ant	0.360	**0.632**	−0.021	−0.112	0.462	0.123	−0.027	−0.105
Flu	**0.913**	0.367	0.003	−0.016	0.113	0.013	0.021	−0.055
Pyr	**0.975**	0.115	0.071	−0.007	−0.027	0.035	−0.047	−0.007
Ba[A]	**0.987**	0.008	0.026	0.064	0.067	0.002	0.057	0.037
Chr	**0.974**	0.058	0.086	−0.001	−0.058	0.052	0.032	−0.042
B[b]F	**0.973**	0.111	0.011	−0.009	0.116	0.020	0.032	0.069
B[k]F	**0.991**	0.061	0.034	0.036	0.041	0.016	0.027	0.018
B[a]P	**0.929**	−0.049	−0.047	0.020	0.133	0.018	0.123	0.128
Ind	**0.986**	0.050	0.004	0.011	0.077	−0.003	0.061	0.041
D[ah]A	**0.971**	0.033	−0.070	0.004	0.033	0.053	−0.006	−0.023
B[ghi]P	**0.940**	0.018	−0.078	−0.039	0.145	−0.052	0.191	0.029
Zn	0.261	−0.091	0.310	**0.613**	**0.563**	−0.159	0.050	0.216
Fe	0.152	−0.158	0.032	0.048	0.133	−0.080	**0.910**	0.015
Al	−0.078	−0.166	**0.933**	−0.056	0.202	0.032	−0.131	0.037
Hg	**0.883**	−0.016	0.013	−0.039	−0.069	0.383	0.044	−0.048
V	0.218	−0.065	**0.750**	0.460	0.108	0.015	0.260	0.099
Cr	0.276	0.188	0.491	0.008	0.133	**0.761**	−0.059	0.100
Mn	0.203	−0.050	0.188	0.314	−0.461	−0.273	0.460	−0.176
Ni	**0.535**	0.016	−0.042	0.203	0.468	−0.004	**0.513**	−0.002
Cu	0.146	0.019	0.153	−0.246	**0.859**	−0.123	0.187	−0.160
As	−0.017	−0.027	0.026	**0.963**	−0.172	−0.031	−0.014	−0.016
Se	0.203	0.298	−0.127	0.112	0.049	−0.299	−0.225	−0.635
Sr	−0.077	0.263	**0.897**	0.177	−0.138	−0.066	0.083	−0.120
Cd	0.123	−0.158	−0.195	−0.033	−0.145	**0.893**	−0.103	−0.039
Ba	−0.184	−0.276	0.357	**0.691**	−0.156	0.070	0.259	−0.174
Pb	0.198	0.154	−0.074	0.022	−0.044	−0.117	−0.130	**0.802**

Concerning PAHs, the first column includes Flut, Pyr, B [a] A, Chr, B [b] F, B [k] F, B [a] P, Ind, D [ah] A and B [ghi] P, while of the metals mainly includes Hg and less Ni. Thus, Factor 1 appears to be related to smoking indoors at home since the aforementioned PAHs belong to the COMPAHs category and are related to combustion processes, while both the Hg and Ni have also been suggested as derivatives of smoking [38]. This is also confirmed by Figure 2, where the concentrations of COMPAHS are considerably high in the houses of smokers compared to those of nonsmokers. Similar results were obtained for Hg (smokers' houses: 0.5, nonsmokers' houses: 0.2 μg/g) and to a lesser extent for Ni (smokers' houses: 24, nonsmokers' houses: 22 μg/g). On Factor 2, the contribution of Acy, Ace, Fl, and Phe is important and to a lesser extent the one of Ant, indicating that their common source is the combustion from vehicle emissions. Therefore, this factor is related to traffic and the aforementioned species originating from ambient air penetrate indoors. On factor 3, the main contribution of Al, followed by Sr and V, indicates that the above elements have mainly natural origin. On factor 4, the main contribution of As and to a lesser extent of Ba and Zn implies the existence of sources related to vehicle traffic, while the presence of Cu and Zn on factor 5 is associated with the road dust resuspension [39]. Regarding factor 6, the significant contribution of Cd and Cr may be attributed to abrasion of plated surfaces (fragments of the coating of metal surfaces or galvanized paints). Finally, on factor 7 the main contribution of Fe indicated that this factor is soil-related, while on factor 8 the great loading of Pb is mainly related to vehicle emissions. The latter is in agreement with the literature since it has been reported that Pb has anthropogenic contamination rather than soil-related origin [31].

3.5. Health Risk Assessment of PAHs and Heavy Metals in Settled House Dust

3.5.1. Cancer Risk Assessment of PAHs in Settled House Dust

A risk assessment using the concentration data of carcinogenic PAHs was conducted in order to assess the potentially detrimental effects associated with exposure to B2PAHs. For the moderate exposure scenario (0.05 g day^{-1}) the results indicated that the risk of carcinogenesis was 1.2×10^{-7}, while for a residence, in which the highest concentrations of B2 PAHs were observed, the risk was 5.4×10^{-7}. These values were found to be one order of magnitude smaller than those of Maertens et al. [18] (1.6×10^{-6}) and Roberts et al. [40] (7.8×10^{-6}). The results of the present study were comparable but slightly higher than those of Langer et al. [7]. For the high exposure scenario (0.1 g day-1), the risk of carcinogenesis was estimated as 2.5×10^{-7} and compared with the values reported in Maertens et al. [18] (3.2×10^{-6}) and Roberts et al. [40] (1.6×10^{-5}) was one and two orders of magnitude lower, respectively. However, in the case of the residence, where the highest concentrations of carcinogenic PAHs were observed, the risk was $1.1 \times 10^{-6,}$ and it was comparable to the one of Maertens et al. [18] and an order of magnitude lower than that of Roberts et al. [40]. Although a strict threshold for risk of carcinogenicity that corresponds to safe exposure has not been established yet, one cancer case per million people (1×10^{-6}) is commonly used as the baseline level of acceptable risk. The values that were obtained by the present study are lower than the above limit for both exposure scenarios. As for the house with the highest levels of exposure, the risk of carcinogenesis for the high exposure scenario marginally exceeded the acceptable risk.

3.5.2. Heavy Metal Risk Exposure in Settled House Dust

Table 4 presents the doses of metals that children and their parents received through all the three routes of exposure, as well as the total dose. According to the results, the ingestion of house dust was found to be the main exposure pathway for metals to children, followed by dermal contact. This finding is in agreement with previously conducted studies [15,41]. The results also indicated that the doses received by the children were one or two orders of magnitude lower than those of their parents. Depending on the magnitude of exposure dose the heavy metals for both children and their parents were sorted as follows: Zn > Cu > Cr > Pb > Ni > Ni > As > Se > Cd > Hg.

Table 4. Exposure to heavy metals in SHD through the different exposure pathways.

Element	$D_{\text{ingestion}}$		$D_{\text{inhalation}}$		D_{Dermal}		D_{total}	
	Child	**Adult**	**Child**	**Adult**	**Child**	**Adult**	**Child**	**Adult**
Cr	1.00×10^{-3}	9.32×10^{-5}	2.11×10^{-8}	1.30×10^{-8}	1.15×10^{-6}	1.40×10^{-7}	1.00×10^{-3}	9.33×10^{-5}
Ni	4.60×10^{-4}	4.27×10^{-5}	9.65×10^{-9}	5.97×10^{-9}	5.29×10^{-7}	6.41×10^{-8}	4.60×10^{-4}	4.27×10^{-5}
Cu	5.22×10^{-3}	4.85×10^{-4}	1.10×10^{-7}	6.78×10^{-8}	6.00×10^{-6}	7.28×10^{-7}	5.22×10^{-3}	4.85×10^{-4}
Cd	8.23×10^{-6}	7.65×10^{-7}	1.73×10^{-10}	1.07×10^{-10}	9.47×10^{-9}	1.15×10^{-9}	8.24×10^{-6}	7.66×10^{-7}
Pb	7.09×10^{-4}	6.58×10^{-5}	1.49×10^{-8}	9.20×10^{-9}	8.15×10^{-7}	9.88×10^{-8}	7.09×10^{-4}	6.59×10^{-5}
Zn	6.16×10^{-3}	5.72×10^{-4}	1.29×10^{-7}	8.01×10^{-8}	7.09×10^{-6}	8.60×10^{-7}	6.17×10^{-3}	5.73×10^{-4}
Hg	5.79×10^{-6}	5.38×10^{-7}	1.22×10^{-10}	$7.53 \times 10^{-}11$	6.66×10^{-9}	8.08×10^{-10}	5.80×10^{-6}	5.39×10^{-7}
As	6.08×10^{-5}	5.64×10^{-6}	1.28×10^{-9}	7.89×10^{-10}	6.99×10^{-8}	8.47×10^{-9}	6.08×10^{-5}	5.65×10^{-6}
Se	1.61×10^{-5}	1.50×10^{-6}	3.39×10^{-10}	2.09×10^{-10}	1.85×10^{-8}	2.25×10^{-9}	1.61×10^{-5}	1.50×10^{-6}

According to Table 5, which presents a comparison between the total doses for children and their parents with the reference doses, the total dose of Cr for children was the same order of magnitude as the reference dose. The doses for all the other elements were found 1–2 order of magnitude lower than the corresponding reference doses. As for the parents, the doses were significantly lower with respect to the children, and the differences between the reference doses were larger (2–4 order of magnitude).

Table 5. Comparison between the total doses for children and their parents with the reference doses.

Element	D_{total}		D_{Ref}
	Child	**Adult**	**Child & Adult**
Cr	1.00×10^{-3}	9.33×10^{-5}	3.0×10^{-3}
Ni	4.60×10^{-4}	4.27×10^{-5}	2.0×10^{-2}
Cu	5.22×10^{-3}	4.85×10^{-4}	2.0×10^{-2}
Cd	8.24×10^{-6}	7.66×10^{-7}	1.0×10^{-3}
Pb	7.09×10^{-4}	6.59×10^{-5}	3.0×10^{-3}
Zn	6.17×10^{-3}	5.73×10^{-4}	3.0×10^{-1}
Hg	5.80×10^{-6}	5.39×10^{-7}	3.0×10^{-4}
As	6.08×10^{-5}	5.65×10^{-6}	3.0×10^{-4}
Se	1.61×10^{-5}	1.50×10^{-6}	5.0×10^{-3}

All HQ values were found to be less than one, and therefore, the risk is limited or even negligible, while as expected all values were found to be higher for children than for adults (Table 6). The element of the house dust associated with the highest noncarcinogenic risk was Cr, for both children and their parents. This finding is in agreement with Shao et al. [27], who also found that Cr was linked with the highest noncarcinogenic risk. On the scales of dangerousness Cu and Pb followed, while Cd was the element that was linked to the lowest risk, probably because it was detected only in low concentrations.

Table 6. Noncarcinogenic exposure risk values for children and their parents.

Element	HQ_{child}	HQ_{adult}
Cr	0.335	0.031
Ni	0.023	0.002
Cu	0.261	0.024
Cd	0.008	0.001
Pb	0.236	0.022
Zn	0.021	0.002
Hg	0.019	0.002
As	0.203	0.019
Se	0.003	0.000

Table 7 presents the lifetime average exposure doses of carcinogenic heavy metals (Cr, Ni, Cd, As) through the inhalation pathway, indicating that the elements are sorted as follows: Cr > Ni > As > Cd. According to the results, the carcinogenic risk exposure dose Exposure Risk was found to be too low to constitute a significant hazard for children and their parents.

Table 7. Carcinogenic risk values of trace metal in inhalation exposure pathway.

Element	LD_{inh}	SF_{inh} ($mg{\cdot}kg^{-1}{\cdot}d^{-1}$)	Exposure Risk
Cr	9.43×10^{-8}	42.00	3.96×10^{-6}
Ni	4.32×10^{-8}	0.84	3.63×10^{-8}
Cd	7.73×10^{-10}	6.30	4.87×10^{-9}
As	5.71×10^{-9}	1.50	8.56×10^{-9}

4. Conclusions

In summary, the dust samples collected from houses of young children in Athens, Greece, are indicative of relatively clean or moderately polluted indoor environments. This is why, the ΣPAH concentrations reported in the present study were in general lower with respect to those recorded in literature, while the metal concentrations were in some cases higher compared to those of previous studies. The calculated EFs of the heavy metals indicated that the elements Cu, Se, Zn, Hg, Cd, and Pb (EF > 40) were mainly of anthropogenic origin. PCA analysis on the chemical composition matrix of house dust showed that the sources that contributed the most to the concentrations of household dust were: smoking, combustion processes, resuspension of soil dust, traffic, and the abrasion of plated surfaces. The cancer risk of PAHs exposure was found lower than the threshold value, with the exception of one residence where high concentrations of carcinogenic PAHs were detected. Health risk assessment indicated that the ingestion of house dust is the most important route of exposure to heavy metals, followed by dermal absorption. As for the children, except of Cr, the doses of all elements were found 1–2 orders of magnitude lower than the corresponding reference doses, and the doses of parents were significantly lower with respect to the children. Regarding the noncarcinogenic risk, the heavy metals were sorted as Cr > Cu > Pb, while the risk of exposure of children and their parents was within the safety limits. Concerning the carcinogenic risk, the heavy metals were sorted as Cr > Ni > As > Cd, while the carcinogenic risk was found too low to be a risk to human health.

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Informed Consent Statement: Informed consent was obtained from all subjects involved in the study.

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