

Article

# Household Dust: Loadings and PM<sub>10</sub>-Bound Plasticizers and Polycyclic Aromatic Hydrocarbons

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**Abstract:** Residential dust is recognized as a major source of environmental contaminants, including polycyclic aromatic hydrocarbons (PAHs) and plasticizers, such as phthalic acid esters (PAEs). A sampling campaign was carried out to characterize the dust fraction of particulate matter with an aerodynamic diameter smaller than 10 µm (PM<sub>10</sub>), using an in situ resuspension chamber in three rooms (kitchen, living room, and bedroom) of four Spanish houses. Two samples per room were collected with, at least, a one-week interval. The PM<sub>10</sub> samples were analyzed for their carbonaceous content by a thermo-optical technique and, after solvent extraction, for 20 PAHs, 8 PAEs and one non-phthalate plasticizer (DEHA) by gas chromatography-mass spectrometry. In general, higher dust loads were observed for parquet flooring as compared with tile. The highest dust loads were obtained for rugs. Total carbon accounted for 9.3 to 51 wt% of the PM<sub>10</sub> mass. Plasticizer mass fractions varied from 5 µg g<sup>-1</sup> to 17 mg g<sup>-1</sup> PM<sub>10</sub>, whereas lower contributions were registered for PAHs (0.98 to 116 µg g<sup>-1</sup>). The plasticizer and PAH daily intakes for children and adults via dust ingestion were estimated to be three to four orders of magnitude higher than those via inhalation and dermal contact. The thoracic fraction of household dust was estimated to contribute to an excess of 7.2 to 14 per million people new cancer cases, which exceeds the acceptable risk of one per million.

**Keywords:** resuspension; household dust; PM<sub>10</sub>; organic and elemental carbon; phthalic acid esters; polycyclic aromatic hydrocarbons

## 1. Introduction

In industrialized nations, people spend most of their time in closed environments, especially at home [1]. Walking induced particle resuspension has been reported to be an important source of indoor particulate matter [2–4]. Several factors affect resuspension of particles, including relative humidity, flooring type, and dust loadings [3]. Household dust is a complex mixture of particles of both indoor and outdoor origin, including organic, inorganic, and biological components, many of which are toxicants, carcinogens or allergens [5,6]. Its composition depends on numerous conditions, such as environmental and seasonal factors, ventilation and air filtration, homeowner activities, and in- and outdoor sources [7]. Several studies have shown that inhalation of dust particles is linked to an increased risk of a range of health hazards, spanning from asthma symptoms in susceptible adults and children [8–11] to cancer and fertility problems [12].

Residential dust is recognized as a major source of environmental contaminants, including polycyclic aromatic hydrocarbons (PAHs) and phthalic acid esters (PAEs) [13–15]. PAHs are primarily byproducts of incomplete combustion of fossil fuels and biomass and pyrosynthesis of organic materials [16]. Dust, especially in carpeted floor, can be a permanent reservoir for these chemicals, which may be inhaled through resuspension into air, ingested accidentally by children or absorbed through the skin [13,17,18]. Because of their widespread sources and strong carcinogenicity, cytotoxicity, mutagenicity, endocrine disrupting, and other hazardous properties, PAHs have been the focus of extensive attention by scientists and governmental organizations [7,18–24]. Benzo[a]pyrene (BaP), the most extensively studied carcinogenic PAH, is classified by the International Agency for Research on Cancer (IARC) as a Group 1 or known human carcinogen [25]. Four of the top ten priority pollutants, nominated by the Agency for Toxic Substances and Disease Registry (ATSDR) in 2011, are single PAHs or PAH mixtures (PAHs, BaP, benzo[b]fluoranthene, and dibenzo[a,h]anthracene) [26].

PAEs (also called phthalates) are used as plasticizers in several consumer products, commodities, and building materials [27]. Therefore, phthalates are ubiquitous in residential and occupational environments, where they are present in high concentrations, both in air and in dust [14,27–41]. Comparisons of mass fractions reported in different studies indicate that, for measurements conducted over the past decade, the levels of PAEs in indoor dust tend to be three to five orders of magnitude higher than those of PAHs [14]. Recent toxicological studies have proven the potential of PAEs to disturb the human hormonal system and human sexual development and reproduction [42,43]. Moreover, PAEs are suspected to trigger asthma and dermal diseases in children ([44], and references therein). An EU risk assessment classified bis(2-ethylhexyl)phthalate (DEHP), dibutyl phthalate (DBP), and benzyl butyl-phthalate (BBP) as hazardous substances in 2005, and has issued a directive to ban these materials from products, particularly toys and cosmetics [45].

Indoor aerosol sources can significantly contribute to the daily dose of particles deposited into the human respiratory system [46]. Since indoor dust contributes to human exposure, its resuspension rates and chemical composition should be evaluated. The selection of appropriate techniques to assess household dust loadings and composition is a major challenge since several methodologies have been employed, including passive (dust settling) and active techniques (surface wiping, press sampling, sweeping, and vacuuming) [6,47].

Studies on the concentration of contaminants in household dust have been focused on the analysis of the total mass or of sieved fractions, involving sizes of tens or hundreds of  $\mu\text{m}$  [17,29,30,32,35,37,39,40]. A summary of average PAH concentrations in settled house dust by country and year (data from 35 studies) can be found in a recent review article [48]. A major issue encountered when comparing these studies was the variability in both the sampling methods employed and the dust particle size fractions subjected to analysis. In 21 out of the 35 studies reviewed, the particle size cut-off points were either 150  $\mu\text{m}$  or 63  $\mu\text{m}$ . It has been suggested that particles  $>150 \mu\text{m}$  do not easily and efficiently adhere to hands or skin. Therefore, these sizes are less relevant when evaluating exposure via ingestion or dermal pathways [49]. Liu et al. [24] collected and sieved indoor dust into six size fractions from office and public microenvironments in Nanjing, China. Higher PAH concentrations, and consequently more health risks, were observed for the smallest particles ( $<43 \mu\text{m}$ ). In general, the common sampling procedures are affected by the loss of fine particles owing basically to the difficulties of collecting all deposited material and to the electrostatic adhesion of particles to brush hairs, vacuum cleaner bags, and sieve meshes. Considering the possibility that household dust is resuspendable and can become airborne, most methodologies have drawbacks when assessing human inhalation exposure. Such an approach requires measurements of the contaminant concentration in smaller particle sizes. The main goal of this study was to determine household dust loadings and the respective PAH and plasticizer levels in the thoracic fraction ( $<10 \mu\text{m}$ ) of resuspendable material on the floor, which deposit anywhere within the lung airways. The in situ resuspension chamber was previously devised and successfully applied to collect the deposited particulate matter with an aerodynamic diameter smaller than 10  $\mu\text{m}$  ( $\text{PM}_{10}$ ) from different road pavements [50,51], but it was the first time this active sampling methodology

was used to collect settled thoracic particles directly from the floor in indoor environments. For road dust, this sampling technique was previously compared to the USEPA methodology (AP-42 documents), which is based on vacuuming or sweeping of surfaces. Then, the collected material passes through a 200-mesh sieve to determine surface silt loadings, and finally an empirical formula is used to estimate PM<sub>10</sub> emission factors. Both methodologies provided very comparable results for resuspendable PM<sub>10</sub> dust [52]. Toxic substances attached to inhaled particles capable of passing beyond the larynx, i.e., PM<sub>10</sub>, could lead to a series of respiratory and cardiovascular diseases, and increase the risks of cancer [53]. Thus, sampling the total concentration or coarser dust fractions most likely provides only a rough estimate of exposure. In addition to household dust loadings and PM<sub>10</sub>-bound chemical components, this paper also provides a relatively exhaustive review of literature values to show not only the order of magnitude of the levels, but also the difficulties in comparing results due to the lack of standard methodologies.

## 2. Methodologies

### 2.1. Sampling

To determine and characterize dust loadings, a sampling campaign was carried out in four different houses located in the Spanish city of León (Table 1). In each housing unit, three rooms were investigated, including the kitchen, the living room and a bedroom. In each room, two samples were collected with, at least, a one-week interval. Samples in each house were taken one or two days before weekly cleaning. For dust collection, an in situ resuspension chamber operating at an air flow rate of 25 L min<sup>-1</sup> was used [51]. After vacuuming, PM<sub>10</sub> was separated from the total dust through a Negretti stainless steel elutriation filter and collected onto 47 mm quartz fiber filters (Pallflex®, Ann Arbor, USA), while particles with aerodynamic diameter >10 µm were deposited in the methacrylate chamber and along the elutriation filter. Electrostatic adhesion could cause some losses of particles <10 µm. However, this loss is likely to be negligible with respect to losses of traditional sampling procedures (i.e., sweeping). In a previous work, the chamber sediments were brought to a laboratory, dried for 48 h at room temperature, sieved and, then, analyzed by means of an optical particle sizer with the aim of verifying the granulometry selection of the sampling system. Results showed that the fraction <10 µm was, on average, only 0.6% and 0.1% (in volume) of samples previously sieved at 250 µm and 63 µm, respectively [50].

**Table 1.** Characteristics of the houses where floor dust particulate matter with an aerodynamic diameter smaller than 10 µm (PM<sub>10</sub>) was sampled.

House	Characteristics	Room	Flooring
1	Suburban two-story house with well-ventilated kitchen, two occupants	Kitchen	Tile
		Bedroom	Parquet
		Living room	Parquet
		Living room rug Bedroom rug	Cut pile carpet/rug Long threads shag rug
2	Single story apartment located in the city center, two occupants	Kitchen	Tile
		Bedroom	Parquet
		Living room	Parquet
3	Rural two-story house with open fireplace in the living room, two occupants	Kitchen	Tile
		Bedroom	Parquet
		Living room	Tile
4	Single story apartment with small kitchen open to the living room, one occupant	Kitchen	Tile
		Bedroom	Tile
		Living room	Tile

Sampling was performed in surface areas of 1 m<sup>2</sup> for 30 min. Two to three different square meters were sampled using the same filter in order to ensure enough particulate mass for the subsequent

gravimetric and chemical analyzes. Since some compounds could derive from the resuspension chamber itself or from ambient air that enters the system and passes through the filter during dust collection from the floor, air samples were vacuumed through the same system after sampling in each room. These background air filters were also sampled for 30 min.

## 2.2. Household Dust Characterization

Due to the loss of a small fragment after sampling, the kitchen filter from house 2 obtained in the 2<sup>nd</sup> week was discarded because weighing led to a negative dust mass. After gravimetric determination, two punches (9 mm) of each filter were analyzed by a thermo-optical transmission technique to obtain the PM<sub>10</sub> carbonaceous content (organic and elemental carbon, OC and EC). This method is based on the quantification of the CO<sub>2</sub> released from the volatilization and oxidation of different carbon fractions under controlled heating by a non-dispersive infrared (NDIR) analyzer. The blackening of the filter is monitored using a laser beam and a photodetector, which enables separating the EC formed by pyrolysis [54]. The remaining portion of each filter was extracted by sonication for 15 min with three aliquots (25 mL each) of dichloromethane. After filtration, the solvent was concentrated in a TurboVap system from Biotage and evaporated to dryness by a gentle nitrogen stream. All the extracts were analyzed by gas chromatography-mass spectrometry (GC-MS) in a Shimadzu QP5050A equipped with a TRB-5MS 30 m × 0.25 mm × 0.25 μm column. The quantitative analysis was performed by single ion monitoring (SIM). Background air filters were analyzed in the same way as the samples to obtain blank-corrected results. Data were acquired in the electron impact (EI) mode (70 eV). The oven temperature program was as follows: 60 °C (1 min), 60 to 150 °C (10 °C min<sup>-1</sup>), 150 to 290 °C (5 °C min<sup>-1</sup>), 290 °C (30 min) and using helium as carrier gas at 1.2 mL min<sup>-1</sup>. For the quantification of PAHs, the following mixture of deuterated internal standards (IS) was used: 1,4-dichlorobenzene-d<sub>4</sub>, naphthalene-d<sub>8</sub>, acenaphthene-d<sub>10</sub>, phenanthrene-d<sub>10</sub>, chrysene-d<sub>12</sub>, and perylene-d<sub>12</sub> (Supelco, St. Louis, USA). In the case of plasticizers, deuterated diethyl phthalate-3,4,5,6-d<sub>4</sub> and bis(2-ethylhexyl)phthalate-3,4,5,6-d<sub>4</sub> (Supelco, St. Louis, USA) were used as IS. Calibrations were performed with authentic standards (Sigma-Aldrich, St. Louis, USA) in eight different concentration levels.

## 2.3. Extraction Recoveries

To assess recoveries during extraction, prebaked blank filters were spiked with known amounts of standards, covering the concentration range commonly reported in the literature for household dust. The solutions of known concentrations were applied to the entire surface of each filter with Pasteur pipettes. The solvent was allowed to evaporate keeping the filters in a desiccator overnight. The filters were subjected to the same methodology of extraction and analysis used for the samples. Six prebaked blank filters were also extracted and analyzed. Four distinct concentrations were tested, in triplicate, and each extract was injected 3 times. The following overall recoveries (%) were obtained: diethyl phthalate 38.4 ± 8.05, di-n-butyl phthalate 113 ± 17.4, benzyl butyl phthalate 70.2 ± 12.7, bis(2-ethylhexyl) adipate 70.4 ± 12.9, bis(2-ethylhexyl) phthalate 108 ± 10.9, phenanthrene 53.0 ± 10.6, anthracene 67.1 ± 11.5, fluoranthene 84.7 ± 11.2, pyrene 81.9 ± 11.0, benzo[a]anthracene 97.2 ± 11.4, chrysene 81.9 ± 9.7, benzo[b]fluoranthene 106 ± 10.0, benzo[k]fluoranthene 108 ± 10.6, benzo[a]pyrene 93.4 ± 13.3, indeno [1,2,3-cd]pyrene 90.6 ± 15.8, dibenzo[a,h]anthracene 87.2 ± 16.7, and benzo[g,h,i]perylene 84.7 ± 16.5. The concentrations shown throughout the manuscript were not adjusted with these percent recoveries. Due to its high volatility and concentration variability in both dust and background air samples, naphthalene was excluded from quantification. Due to its vapor pressure of 0.087 mm Hg at 25 °C, naphthalene is sometimes considered a “borderline” volatile/semivolatile compound, since it may often be detected in both gas and particulate phases. Because of its tendency to sublime, in ambient air, naphthalene is known to mainly exist in the vapor phase [55].

#### 2.4. Health Risk Evaluation

Human exposure to plasticizers occurs via ingestion, inhalation, and dermal contact. The daily intake (DI) through different pathways were estimated by the following equations ([39], and references therein), where  $DI_{ing}$ ,  $DI_{inh}$  and,  $DI_{der}$  are the daily intakes ( $ng\ kg^{-1}\ day^{-1}$ ) via dust ingestion, inhalation, and dermal contact, respectively:

$$DI_{ing} = \frac{C_{dust} \times IngR \times EF \times ED \times CF}{BW \times AT} \quad (1)$$

$$DI_{inh} = \frac{C_{dust} \times InhR \times EF \times ED}{BW \times AT \times PEF} \quad (2)$$

$$DI_{der} = \frac{C_{dust} \times SA \times AF_{dust} \times ABS \times EF \times ED \times CF}{BW \times AT} \quad (3)$$

$C_{dust}$  represent the mean concentrations in dust ( $ng\ g^{-1}$ );  $IngR$  is the ingestion rate of indoor dust ( $200\ mg\ day^{-1}$  for children,  $100\ mg\ day^{-1}$  for adults);  $EF$  is the exposure frequency (180 days per year for both children and adults);  $ED$  is the exposure duration (6 years for children, 24 years for adults);  $CF$  is an unit conversion factor ( $10^{-3}\ g\ mg^{-1}$ );  $BW$  is the body weight (15 kg for children, 70 kg for adults);  $AT$  is the average time (2190 days for children, 8760 days for adults);  $InhR$  is the inhalation rate ( $7.6\ m^3\ day^{-1}$  for children,  $12.8\ m^3\ day^{-1}$  for adults);  $PEF$  is the particulate emission factor ( $1.36 \times 10^6\ m^3\ g^{-1}$ );  $SA$  is the dermal exposure area ( $1150\ cm^2$  for children,  $2145\ cm^2$  for adults);  $AF_{dust}$  is the dust adherence factor ( $0.2\ mg\ cm^{-2}\ day^{-1}$  for children,  $0.07\ mg\ cm^{-2}\ day^{-1}$  for adults); and  $ABS$  is the dermal adsorption fraction (0.001 for both children and adults, dimensionless).

Like plasticizers, PAHs can enter the body through ingestion (swallowing), inhalation (breathing), and skin contact. The carcinogenic potency of PAHs in indoor dust can be evaluated by comparing the carcinogenic potency of each PAH to that of BaP. The BaP carcinogenic equivalent concentration ( $BaP_{TEQ}$ ) is defined as follows:

$$BaP_{TEQ} = \sum C_i \times BaP_{TEF} \quad (4)$$

where  $C_i$  is the concentration of each individual PAH and  $BaP_{TEF}$  are toxic equivalency factors [24]. The incremental lifetime cancer risk (ILCR) is generally used to quantitatively estimate the exposure risks from the three exposure routes [24,56]:

$$ILCR_{ing} = \frac{CS \times (CSF_{ing} \sqrt[3]{BW/70}) \times IngR \times EF \times ED \times CF}{BW \times AT} \quad (5)$$

$$ILCR_{inh} = \frac{CS \times (CSF_{inh} \times \sqrt[3]{BW/70}) \times InhR \times EF \times ED}{BW \times AT \times PEF} \quad (6)$$

$$ILCR_{der} = \frac{CS \times (CSF_{der} \times \sqrt[3]{BW/70}) \times SA \times AF_{dust} \times ABS \times EF \times ED \times CF}{BW \times AT} \quad (7)$$

where  $CS$  is the  $BaP_{TEQ}$  concentration ( $mg\ kg^{-1}$ );  $CSF_{ing}$ ,  $CSF_{inh}$ , and  $CSF_{der}$  are carcinogenic slope factors of 7.3, 3.85, and 25 ( $mg\ kg^{-1}\ day^{-1}$ )<sup>-1</sup>, respectively.

The risk associated with non-carcinogenic PAHs in household  $PM_{10}$  dust was estimated through the hazard quotient (HQ):

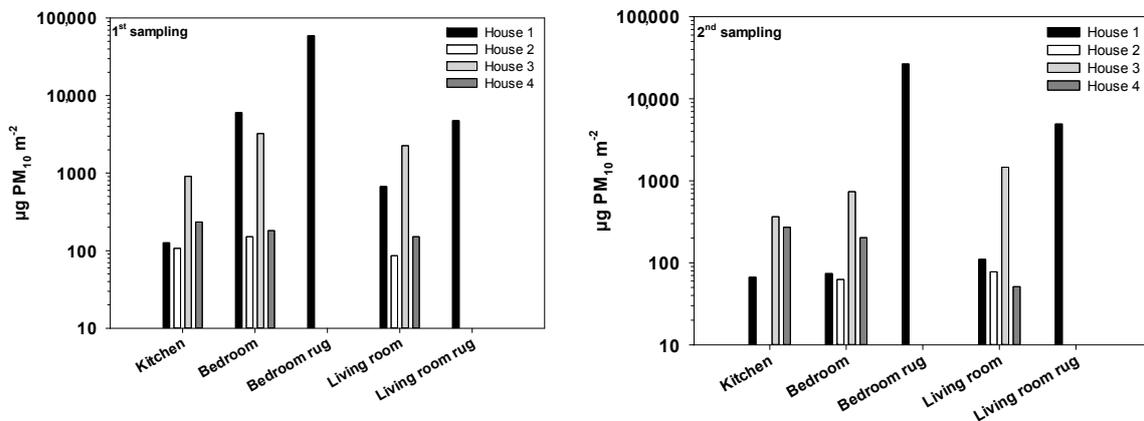
$$HQ = DI_{nc}/R_fD \quad (8)$$

where  $DI_{nc}$  is the daily intake via ingestion of non-carcinogenic PAHs, obtained by Equation (1), and  $R_fD$  is the reference dose. The oral ingestion  $R_fD$  values were taken from Iwegbue et al. [57].

### 3. Results and Discussion

#### 3.1. Dust Loadings

Huge differences in dust loadings between rugs and hard floorings were registered (Figure 1). Rugs represented the surface with the highest dust loadings. In general, higher dust loadings were observed for parquet flooring as compared with tile. Among the four dwellings, the highest dust loadings were obtained in the living room of a suburban family house with an open fireplace. Several factors can affect the surface dust loadings, such as walking, cleaning frequency, household materials, and indoor particle sources. Thus, the presence or absence of occupants can have a marked effect on resuspension levels. In house 1, for example, a very significant decrease in dust loadings from the first to the second week was registered, possibly due to the absence of owners for a few days. The highest mean value was observed in bedrooms ( $1745 \mu\text{g PM}_{10} \text{ m}^{-2}$ ), followed by living rooms ( $785 \mu\text{g PM}_{10} \text{ m}^{-2}$ ) and, finally, kitchens ( $297 \mu\text{g PM}_{10} \text{ m}^{-2}$ ). The size distribution of aerosols emitted from cooking activities has been reported to be dominated by ultrafine particles, with modes generally in the range of 20–100 nm [58]. Thus, although cooking emissions can be high and contribute to the deposition and subsequent resuspension of dust, nanometer sizes represent a small fraction of the particulate mass. Dust loadings in the kitchen of house 3 were two to seven times higher than in the kitchens of other houses. This may be due to a higher utilization rate. In the other three residences the owners, in general, only prepare dinner. A higher mean value for bedrooms may be related to the presence of specific active sources or activities in this space of the house, such as making and unmaking the bed, dressing and undressing (which potentiates skin desquamation), existence of various rugs, use of cosmetics, etc. [59].



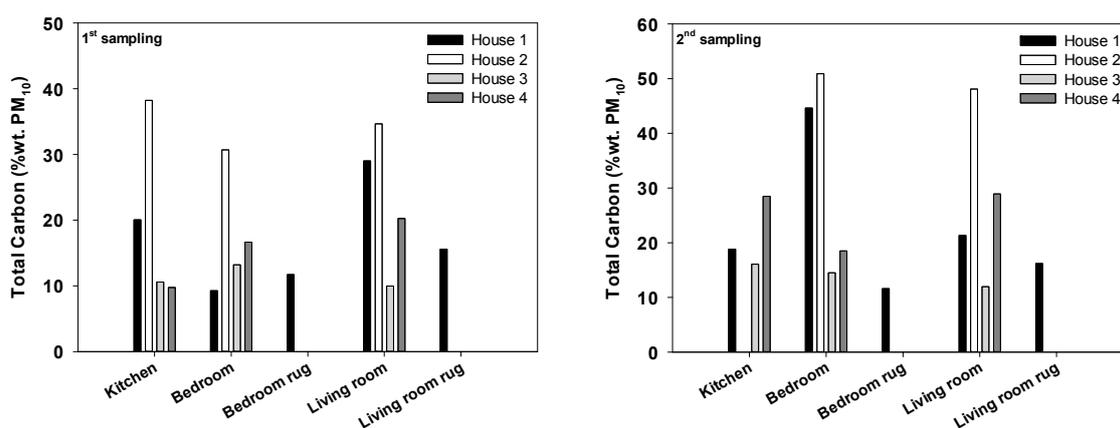
**Figure 1.** Dust loadings in the different rooms of the various houses for the two sampling campaigns.

Using a mechanical resuspension device, Tian et al. [3] characterized walking-induced particle resuspension as a function of flooring type. Results showed that for particles at 0.4 to 3.0  $\mu\text{m}$ , the difference in resuspension fraction between carpets and hard floorings was not significant. It was also found that for fine particles (0.4 to 3.0  $\mu\text{m}$ ), the difference in resuspension caused by flooring type is negligible, while for coarse particles (3.0 to 10  $\mu\text{m}$ ) carpets are associated with two to four times higher resuspended concentration in comparison with hard floorings. In fact, carpeted floors may contribute to significantly higher surface dust loadings and allergen concentrations than hard floors [60–64]. Roberts et al. [65] used a high-volume surface sampler to measure surface dust in carpets. Dust loadings ranged from 0.32 to 14.4  $\text{g m}^{-2}$ . Adgate et al. [66] collected bare floor and carpet dust samples in 216 Jersey City, New Jersey, homes using quantitative wipe and vacuum sampling techniques. Dust loadings varied from 0.05 and 7.0  $\text{g m}^{-2}$  and from 0.3 and 99  $\text{g m}^{-2}$  in the wipe floor and vacuum samples, respectively. It should be noted that dissimilar indoor dust sampling strategies (e.g., wipe versus vacuum methods) are used to measure loadings and the amounts of

toxicants per unit area, which renders comparisons between studies difficult. Lioy et al. [67] found that while loadings were substantially greater with wipe sampling, metal concentrations within the dust samples were similar for both methods of sampling. Vacuum cleaner sampling has its own series of problems, especially the variability in design and efficiency, and likely does not retain particles below 10  $\mu\text{m}$  [47]. Bai et al. [68] evaluated the following five methods of sampling lead-contaminated dust on carpets: (i) wipe, (ii) adhesive label, (iii) C18 sheet, (iv) vacuum, and (v) hand rinse. The wipe and vacuum methods showed the best reproducibility and correlation with other sampling techniques. The authors concluded that surface wipe sampling was the best method to measure accessible lead from carpets for exposure assessment, while vacuum sampling was most effective for providing information on total lead accumulation (long-term concentrations). In their review paper, Lioy et al. [6] stated, “although we have come a long way in determining the uses of house dust to identify sources of indoor contamination and to provide improved estimates of residential human exposure, one of the challenges still lies in the reliability of sampling techniques.” The instrument used in this study has the advantage of being a device capable of collecting dust particles below 10  $\mu\text{m}$ , for gravimetric and chemical analysis. Sampling and analysis of higher dust particle sizes may only provide a crude estimate of inhalation exposure. However, validation and intercomparison studies are still needed, which is not easy, as the panoply of established methodologies does not match inhalable sizes.

### 3.2. Carbonaceous, Plasticizer, and PAH Particulate Mass Fractions

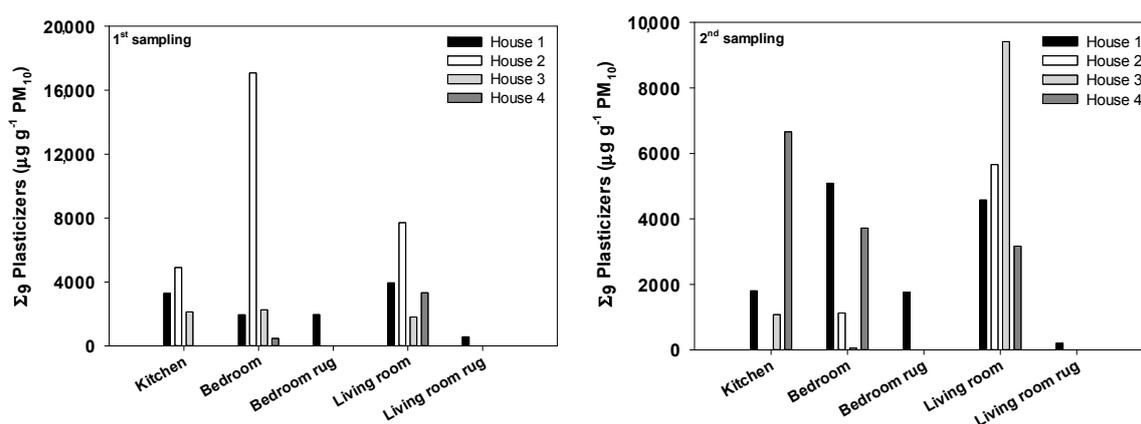
Total carbon accounted for 9.3 to 51 wt% of  $\text{PM}_{10}$  with the highest mass fractions recorded in dust samples collected in the city center apartment (Figure 2). More than 80% of the total carbonaceous matter was composed of OC, whereas in many samples the EC was too low or undetectable. The highest percentages of EC (10% to 20% of total carbon) were observed in kitchens, where there are sources of combustion. The high proportion of OC reflects the importance of a multitude of indoor sources that contribute to the organic carbonaceous component of household dust, including bacteria, skin flakes, cosmetics, cleaning products, cooking, paper and clothing fibers, microscopic specks of plastics, environmental contaminants brought on the soles of our shoes, etc. Our finding that a large portion of  $\text{PM}_{10}$  from indoor dust is composed of OC agrees with the results of Polidori et al. [69], who measured particulate OC and EC concentrations at 173 homes in the USA. These authors demonstrated that part of the OC can be secondarily formed in the indoor environment as a result of reactions involving gas-phase organic compounds emitted by cleaning products, air fresheners, and other sources.



**Figure 2.** Total carbon (TC = OC + EC) mass fraction in resuspendable  $\text{PM}_{10}$  from household dust.

Eight phthalate plasticizers (PAEs) and one non-phthalate plasticizer [bis(2-ethylhexyl) adipate, DEHA] were quantified in  $\text{PM}_{10}$  from settled house dust. PAEs included dimethyl phthalate (DMP), dimethylpropyl phthalate (DMPP), diethyl phthalate (DEP), diisobutyl phthalate (DIBP), di-n-butyl phthalate (DBP), di-n-hexylphthalate (DNHP), benzyl butyl phthalate (BBP), dicyclohexyl phthalate

(DCHP), bis(2-ethylhexyl) phthalate (DEHP), di-n-octyl phthalate (DNOP), di-isononylphthalate (DINP), and di-isodecylphthalate (DIDP). The total mass fractions varied from  $5 \mu\text{g g}^{-1}$  to  $17 \text{mg g}^{-1}$   $\text{PM}_{10}$  (Figure 3). As observed with dust loads, huge differences were observed from week to week and from home to home. The highest values were registered in the bedroom of the city center apartment and in the living room of the rural house. In addition to variability in sources, the concentrations of these compounds depend on the same factors already mentioned for the dust loads (e.g., ventilation, domestic routines, and cleaning activities). Widely scattered concentration levels were also documented in many previous works. For example, Kubwabo et al. [33] analyzed 17 PAEs in 126 Canadian household dust samples, evidencing the huge variability in spatial and temporal distribution of these compounds across different areas of the home, and thus the difficulty in predicting potential household exposures.



**Figure 3.** Plasticizer mass fractions in  $\text{PM}_{10}$  from resuspended household dust.

Most likely due to its high volatility, DMP was the compound with the lowest mass fractions (Table 2). On the other hand, DEHP, DNOP, and DBP were the major PAEs in household dust. While high DBP values were observed in all parquet floor bedrooms, only samples from two living rooms showed detectable masses. Bamai et al. [70] also associated higher DBP levels in floor dust with compressed wooden floor. This type of flooring is usually composed of thin pieces, which are glued together and covered with wax, paint, and sometimes flame retardants. The surface applied products (gloss agents, plastic additives, paint, and varnish) contain DBP [70]. From quantitative and qualitative emission data on phthalates from different materials, Afshari et al. [71] reported that polyolefin covered with wax for floor polishing increased DBP concentration in chamber air by two-fold. DBP is also employed as a coalescing aid in latex adhesives, as well as a plasticizer in cellulose plastics and a solvent for dyes [70]. Furthermore, DBP has been reported to be largely present in cosmetic and personal care products [72].

Although the concentration of DNOP in floor dust has been reported in only a very limited number of publications, the mass fractions of this study are higher than those described in the literature. There were no appreciable differences between the amounts found in  $\text{PM}_{10}$  of the various rooms of the houses. DNOP is used in carpet back coating, floor tile, and adhesives. It is also employed in cosmetics and pesticides [73]. DEHP was present at higher concentrations in the bedroom and living room samples. Although DEHP has been consistently described as one of the most abundant PAEs in settled dust, the levels have decreased over time, reflecting its phase-out in the EU [28]. In Europe, the use of DEHP decreased drastically in 2001 and has to a large extent been replaced by DINP and DIDP, with their longer chains and lower volatility [74]. DEHP has been used in numerous consumer products, children toys, medical devices, and building materials (e.g., vinyl flooring, furniture, paints, cables, wires, wall coverings, and packaging materials) [75]. Bamai et al. [70] and Bornehag et al. [34] associated polyvinyl chloride (PVC) flooring with DEHP levels in house dust. Kolarik et al. [76] documented higher concentrations of BBP, DNOP, and DEHP in indoor dust in homes where polishing

agents were employed as compared with homes where such products were infrequently used or not used at all. As PVC flooring was not present in any of the houses for this study, other emission sources may have contributed to the DEHP levels. It has been shown that the source characteristics (surface area and material phase concentration of DEHP), as well as the external mass-transfer coefficient and ventilation rate, are important variables that influence the steady-state DEHP concentration and the resulting exposure [77]. DEHP and other PAEs are strongly sorbed to surfaces. A relatively small gas-phase concentration, such as 0.1 ppb, is enough for significant vapor transport of a PAE and its subsequent partitioning between the gas phase and indoor surfaces, including airborne particles and settled dust [78].

BBP was present in all samples at relatively similar concentrations regardless of room type. The values fall within the range reported for household dust from other regions. BBP is commonly employed as a plasticizer for vinyl foams, which are often used as floor tiles. Other uses are in artificial leather, paints, and adhesives. BBP was classified as toxic by the European Chemical Bureau, and therefore its use has decayed rapidly in the last decade [79].

No appreciable differences were found between DINP concentrations in samples from the various rooms of the houses. DIPD, instead, was present in higher amounts in living rooms and at lower levels in kitchens. The values observed in this study for these two compounds seem to be lower than those documented by Santillo et al. [80] for dust samples of houses in several European cities, although the ranges reported by these researchers are very wide. On the basis of a risk assessment, in 2013, the European Chemicals Agency (ECHA) concluded that there is no evidence that would justify a re-examination of the existing restriction on DINP and DIDP in toys and childcare articles which can be placed in the mouth by children [81]. About 95% of DINP is used in PVC applications. The other 5% is employed in non-PVC applications such as rubbers, adhesives, sealants, paints and lacquers, and lubricants. For DIDP, non-PVC applications are comparatively small, but comprise use in anticorrosion and antifouling paints, sealing compounds and textile inks ([81], and references therein).

**Table 2.** Comparison of mass fractions of plasticizers ( $\mu\text{g g}^{-1}$ ) in house dust.

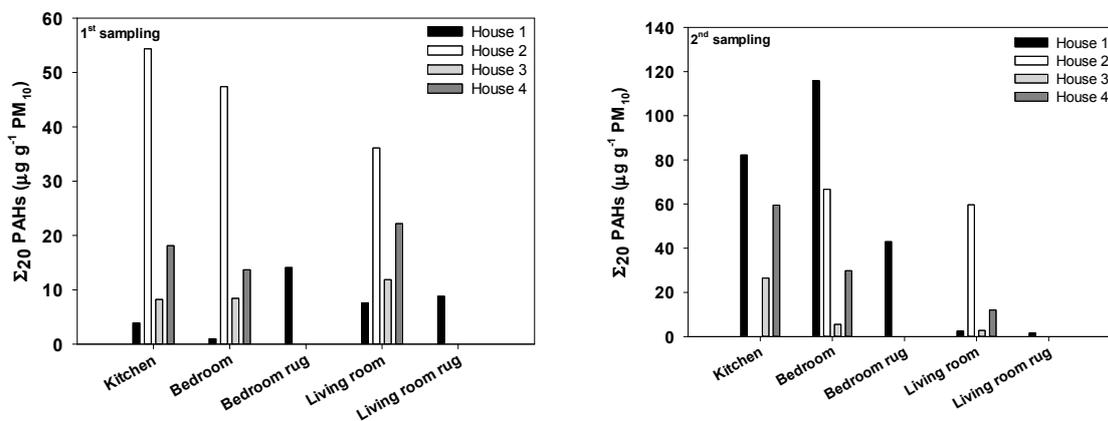
Sampling Information	Statistics	DMP	DMPP	DEP	DIBP	DBP	DNHP	BBP	DEHA	DCHP	DEHP	DNOP	DINP	DIDP	Ref.
Kitchens	Mean	0.27		142		562		6.3	17		271	927	10	6.6	This study
	min-max	<0.1–1.89		<0.1–567		<0.1–3301		<0.1–12	<0.1–97		<0.1–795	<0.1–3270	<0.1–25	<0.1–46	
	Median	<0.1		<0.1		<0.1		6.4	<0.1		<0.1	407	9.0	<0.1	
Bedrooms	Mean	0.33		45		2353		10.4	36		897	969	20	35	This study
	min-max	<0.1–1.57		<0.1–354		<0.1–9397		4.25–26	<0.1–191		<0.1–2864	<0.1–2594	4.15–78	<0.1–215	
	Median	0.19		<0.1		964		7.0	7.4		442	316	7.3	3.9	
Living rooms	Mean	0.93		139		1181		11	19		1352	751	28	66	This study
	min-max	<0.1–4.5		<0.1–924		<0.1–6044		5.1–21	<0.1–109		<0.1–8374	<0.1–2468	<0.1–145	<0.1–300	
	Median	<0.1		16		<0.1		9.3	<0.1		218	473	8.2	16.6	
Living room rug *	Mean	0.60		21		893		8.54	23.4		1997	807	9.8	23.8	This study
Bedroom rug *	Mean	1.31		5.8		139		3.44	3.7		92	119	5.3	9.1	This study
Dust samples (<180 $\mu\text{m}$ ) from houses of China collected with brushes	Mean	0.693		0.187	17.1	26.4	0.003	0.040		0.015	105	0.342			[39]
	Median	0.181		0.116	9.33	12.9	0.001	0.014		0.007	25.6	0.130			
30 household dust samples (<63 $\mu\text{m}$ ) from vacuum cleaner	Median					87.4		15.2			604		129	33.6	[32]
Dust from 30 apartments in bags of vacuum cleaner	Mean	10.8	54.6	44.6		55.6		86.1			776				[82]
	Median	1.5	37.5	6.1		47.0		29.7			703				
Dust (<250 $\mu\text{m}$ ) from 11 houses, 3 labs and 1 hospital in vacuum cleaner bags, Qatar	Mean	0.89		7.9		12.5	1.6	2.6	10.7		288	5.8	106	11.4	[35]
	Median	0.98		7.0		17.0	0.12	1.95	9.6		395	3.0	101	11.0	
Dust samples (<250 $\mu\text{m}$ ) from homes in Kuwait from vacuum cleaner bags	Mean	0.01		1.5		51	0.2	6.4		0.3	1700	14			[83]
	Median	0.03		1.8		45	0.39	8.6		2.9	2256	14			
House dust from China collected by sweeping the floor and wiping the top of furniture (<2 mm)	Median	0.2		0.4	17.2	20.1	nd	0.2		nd	228	0.2			[40]
Dust from Albany, USA, from vacuum cleaner bags of several homes (<2 mm)	Median	0.08		2.0	3.8	13.1	0.6	21.1		nd	304	0.4			[40]
Saudi floor dust from vacuum cleaner bags (<250 $\mu\text{m}$ )	Mean	1.4		4.2	33.6	80.2		1.5			1140	102.4			[37]
	Median	0.6		1.4	22.1	33.3		0.8			1020	26.8			
Kuwaiti floor dust from vacuum cleaner bags (<250 $\mu\text{m}$ )	Mean	0.2		5.1	20.0	4.0		1.3			220	2.4			[37]
	Median	0.1		2.7	17.2	1.6		0.8			240	2.8			

Table 2. Cont.

Sampling Information	Statistics	DMP	DMPP	DEP	DIBP	DBP	DNHP	BBP	DEHA	DCHP	DEHP	DNOP	DINP	DIDP	Ref.
Settled dust collected in child's room, above the floor level, in Bulgarian homes by vacuum cleaner	Mean	260		350		7850		320			960	250			[76]
	Median	280		340		9930		340			1050	300			
Dust from rooms in Germany collected with vacuum cleaner (<2 mm)	Mean	1.32		80.7	107.1	384		85.8			1026		114	13.5	[80]
	Median	1.42		12.9	36.5	44.1		82.2			996		113	< 0.1	
Dust from rooms in Spain (Madrid) collected with vacuum cleaner (<2 mm)	Mean	0.14		14.0	265	131		6.55			464		69.5	10.2	[80]
	Median	< 0.1		7.88	201	145		5.3			370		<0.1	<0.1	
Dust from rooms in France (Paris) collected with vacuum cleaner (<2 mm)	Mean	<0.1		9.88	91.4	174		227			111		122	48.2	[80]
	Median	<0.1		8.72	86.1	65.8		200			356		115	<0.1	
Dust from rooms in Italy (Rome) collected with vacuum cleaner (<2 mm)	Mean	0.30		9.60	221	36.6		89.0			503		76.0	142	[80]
	Median	<0.1		6.78	180	42.8		23.6			434		<0.1	<0.1	
Dust from rooms in the UK collected with vacuum cleaner (<2 mm)	Mean	0.12		12.2	52	50.2		56.5			192		48.5	20.8	[80]
	Median	<0.1		3.5	43.2	52.8		24.5			195		<0.1	<0.1	
Settled dust from apartments in Stockholm from vacuum cleaner	Median	0.47		14	104	103		16			449	0.00	106	56	[74]
Dust from the floor surface and from objects within 35 cm above the floor with vacuum cleaner in Japanese dwellings	Median	<0.5		< 0.5	3.1	16.6		2.0	8.0		1110		139		[70]
Floor dust samples collected with vacuum cleaner from living rooms in 41 dwellings, Sapporo (Japan)	Median	<0.2		0.33	2.9	19.8		4.2	6.5		880		126		[84]
Dust samples from household vacuum cleaner bags in French dwellings (<100 µm)	Mean	0.26		10.6	111	10.5		25.8			441		158		[17]
	Median	0.25		4.9	20	9.1		6.1			462		139		
House dust from urban dwellings in Nanjing, China, collected with vacuum cleaner (150 µm)	Mean	0.4		0.9		52.3		2.9			462	1.6			[36]
	Median	0.1		0.2		23.7		1.6			183	0.1			
Dust from homes across the USA with vacuum cleaner (<1.4 mm)	Mean	0.05		2.34	6.24	4.17		214	77.1		97.2	144			[29]
	Median	0.04		0.49	4.08	3.54		51.2	21.8		73.1	74.9			

\* only two samples, dimethyl phthalate (DMP), dimethylpropyl phthalate (DMPP), diethyl phthalate (DEP), diisobutyl phthalate (DIBP), di-n-butyl phthalate (DBP), di-n-hexylphthalate (DNHP), benzyl butyl phthalate (BBP), bis(2-ethylhexyl) adipate (DEHA), dicyclohexyl phthalate (DCHP), bis(2-ethylhexyl) phthalate (DEHP), di-n-octyl phthalate (DNOP), di-isononylphthalate (DINP), and di-isodecylphthalate (DIDP); nd, not detected.

As compared with plasticizer compounds, PAHs accounted for a much smaller mass of household dust ( $\Sigma_{20}$ PAHs 0.98 to 116  $\mu\text{g g}^{-1}$   $\text{PM}_{10}$ ). Given the small number of samples and the variability in concentrations, it is difficult to infer patterns between house or room typologies (Figure 4). The values obtained in this study fall into the broad range of values reported in the literature. Wang et al. [22] analyzed 15 PAHs in settled house dust of urban dwellings with preschool-aged children in Nanjing, China.  $\Sigma_{15}$ PAHs ranged from 1.2 to 280.4  $\mu\text{g g}^{-1}$ , averaging 11.1  $\mu\text{g g}^{-1}$ . Yadav et al. [85] investigated the contamination level of EPA's priority PAHs in indoor dust from residential, educational, commercial, public places, and office premises, in four major cities of Nepal. Concentrations of  $\Sigma_{16}$ PAHs ranged from 747 to 4910  $\text{ng g}^{-1}$  (median 1320  $\text{ng g}^{-1}$ ). In Palermo, Italy, Mannino and Orecchio [86] collected indoor dust samples by brushing from surfaces at a height of 1.5 to 2.0 m above ground level in bedrooms, living rooms, kitchens, laboratories, offices, in a market, and in a car. The  $\Sigma_{16}$ PAH concentrations were within a broad interval (36 to 34,453  $\mu\text{g g}^{-1}$ ), with an average of 5111  $\mu\text{g g}^{-1}$ , indicating heterogeneous levels of contamination in the investigated microenvironments. Organic extracts of sieved vacuum cleaner dust from 51 homes in Canada were examined for the presence of 13 PAHs [87]. Total concentrations varied between 1.5 and 325  $\mu\text{g g}^{-1}$  with a geometric mean of 12.9  $\mu\text{g g}^{-1}$ . These values were found to be comparable to those documented in a previous review in which the total PAHs in samples collected from urban, rural, and suburban homes ranged between 0.4–544  $\mu\text{g g}^{-1}$  with a geometric mean of 4.5  $\mu\text{g g}^{-1}$  [7]. High concentrations of  $\Sigma_{16}$ PAHs were also observed in indoor dust samples collected across China from 45 private domiciles and 36 public buildings (1.00 to 470  $\mu\text{g g}^{-1}$ , mean value of 30.9  $\mu\text{g g}^{-1}$ ) [88]. It must be noted, once again, that comparability between results of various works should be made with caution, as they concern different surfaces, particle sizes, sampling and analytical methodologies, and list of compounds.



**Figure 4.** Polycyclic aromatic hydrocarbon (PAH) mass fractions in  $\text{PM}_{10}$  dust samples collected in two different sampling periods.

Low molecular weight-PAHs (LMW, two and three rings) were less abundant than high molecular weight-PAHs (HMW, four and six rings), suggesting the dominance of pyrogenic sources. Regardless of the microenvironment, the median LMW/HMW ratios were always in the range from 0.3 to 0.5. The main PAHs in the thoracic fraction of resuspended dust were pyrene, retene, and indeno[1,2,3-cd]pyrene, reaching concentrations up to 27, 21, and 13  $\mu\text{g g}^{-1}$ , respectively (Table 3). While the medians of the latter two compounds were higher in the  $\text{PM}_{10}$  sampled in the living rooms, pyrene showed higher levels in the kitchens. PAH levels and speciation are highly dependent on the cooking methods [58], biomass burning appliances and operating conditions [89], traffic fleet and meteorology in the outdoor surrounding environment [90], among other factors.

**Table 3.** Comparison of PAH levels ( $\mu\text{g g}^{-1}$ ) in household dust from different countries.

PAHs	This Study								[91]	[85]	[87]	[88]	[92]					
	Kitchens		Living Rooms		Bedrooms		Rug Bedroom	Rug Living Room	House Vacuum Samples Brisbane	Indoor Dust from Distinct Buildings Nepal	Settled Dust from Homes in Ottawa		Dust Samples from Private Domiciles and Public Buildings, China		Saudi Household Floor Dust		Kuwaiti Household Floor Dust	
	Mean Min-Max	Median	Mean Min-Max	Median	Mean Min-Max	Median	Mean	Mean	Mean	Median	Mean	Median	Mean	Median	Mean	Median	Mean	Median
ACY	1.75 <0.001–7.89	0.432	3.22 <0.001–11.8	1.15	4.70 0.125–13.0	2.04	0.988	<0.001	6.92	0.031	0.039	0.005	0.173	0.055	0.160	0.080	0.090	0.060
ACE	0.344 <0.001–1.50	<0.001	0.74 <0.001–2.81	0.215	0.246 <0.001–1.45	0.063	0.067	0.136	202	0.018			0.093	0.052	0.110	0.105	0.065	0.060
FLU	0.073 <0.001–0.349	<0.001	0.036 <0.001–0.287	<0.001	0.017 <0.001–0.081	<0.001	0.036	0.012	12.6	0.048	0.170	0.093	0.464	0.200	0.290	0.170	0.165	0.080
PHE	0.213 <0.001–0.876	<0.001	0.260 <0.001–0.987	0.154	0.306 <0.001–1.16	0.177	0.169	0.275	8.77	0.173	2.78	1.48	4.19	1.67	0.425	0.160	0.510	0.410
ANT	1.56 <0.001–10.0	<0.001	1.07 <0.001–8.57	<0.001	< 0.001 <0.001–0.003	<0.001	0.002	1.52	3.61	0.062	0.485	0.196	0.418	0.196	0.105	0.050	0.032	0.025
FLUA	1.56 <0.001–8.48	0.277	1.12 <0.001–4.58	<0.001	1.60 <0.001–11.4	0.045	<0.001	0.197	53.6	0.128			4.26	1.77	0.425	0.175	0.220	0.140
PYR	4.46 <0.001–19.3	2.17	3.67 <0.001–11.2	1.51	4.65 <0.001–27.0	0.604	0.003	1.06	77.3	0.107			2.89	1.18	0.385	0.160	0.035	0.017
CHR	0.921 <0.001–3.65	0.404	0.695 <0.001–2.36	0.506	0.610 <0.001–3.41	0.23	0.120	0.053	5.42	0.157	3.29	1.19	1.79	0.717	0.095	0.085	0.055	0.040
PER	4.29 <0.001–22.4	<0.001	3.73 <0.001–16.8	1.70	3.26 0.001–14.3	1.25	0.301	0.602										
CAR	1.08 <0.001–7.89	<0.001	1.83 <0.001–7.76	0.440	1.64 <0.001–11.6	<0.001	0.001	0.141										
TER	<0.001	<0.001	0.135 <0.001–0.765	<0.001	0.021 <0.001–0.106	<0.001	0.028	<0.001										
RET	3.15 <0.001–15.4	0.737	5.02 <0.001–16.2	2.20	5.28 <0.001–21.4	0.966	<0.001	2.09										
BaA	1.55 <0.001–3.60	1.78	0.516 <0.001–1.67	0.136	0.333 <0.001–2.38	<0.001	0.040	1.40	6.39	0.078	2.38	0.696	2.37	0.569	0.090	0.080	0.090	0.055

Table 3. Cont.

PAHs	This Study								[91]	[85]	[87]	[88]	[92]					
	Kitchens		Living Rooms		Bedrooms		Rug Bedroom	Rug Living Room	House Vacuum Samples Brisbane	Indoor Dust from Distinct Buildings Nepal	Settled Dust from Homes in Ottawa		Dust Samples from Private Domiciles and Public Buildings, China		Saudi Household Floor Dust		Kuwaiti Household Floor Dust	
	Mean Min-Max	Median	Mean Min-Max	Median	Mean Min-Max	Median	Mean	Mean	Mean	Median	Mean	Median	Mean	Median	Mean	Median	Mean	Median
BbF	1.91 <0.001–6.68	1.63	2.10 <0.001–5.28	1.31	1.50 <0.001–6.21	0.124	0.304	1.07	11.3	0.018	4.87	1.66	6.23	1.52	0.700	0.575	0.245	0.210
BkF	1.10 <0.001–5.56	<0.001	1.02 <0.001–3.51	<0.001	0.960 <0.001–4.84	<0.001	<0.001	0.710	3.48	0.075	1.60	0.582	0.763	0.248	0.110	0.105	0.070	0.040
BeP	0.206 <0.001–1.44	<0.001	0.844 <0.001–4.50	<0.001	<0.001	<0.001	<0.001	0.746										
BaP	1.53 <0.001–4.91	1.63	1.23 <0.001–3.27	0.564	1.11 <0.001–3.43	0.219	0.061	1.43	2.97	0.094	2.91	0.803	1.71	0.310	0.550	0.290	0.140	0.080
IcdP	3.48 0.170–12.9	1.75	3.11 0.979–5.13	2.85	2.76 <0.001–8.29	2.32	2.61	0.565	3.34	0.115	3.07	0.911	1.97	0.356	0.015	<0.001	0.055	<0.001
DahA	0.289 0.290–1.22	<0.001	0.790 <0.001–4.31	0.314	0.395 <0.001–1.52	0.155	0.162	0.071	20.1	0.046	0549	0.185	0.510	0.090	0.016	<0.001	0.015	<0.001
BghiP	2.43 <0.001–13.2	0.922	1.10 <0.001–4.40	0.275	1.46 <0.001–6.40	<0.001	<0.001	0.263	6.08	0.068	2.79	0.793	2.14	0.398	0.105	0.075	0.070	<0.001

ACY, acenaphthylene; ACE, acenaphthene; FLU, fluorene; PHE, phenanthrene; ANT, anthracene; FLUA, fluoranthene; PYR, pyrene; CHR, chrysene; PER, perylene; CAR, carbazole; TER, p-terphenyl; RET, retene; BaA, benzo[a]anthracene; BbF, benzo[b]fluoranthene; BkF, benzo[k]fluoranthene; BeP, benzo[e]pyrene; BaP, benzo[a]pyrene; IcdP, indeno[1,2,3-cd]pyrene; DahA, dibenzo[a,h]anthracene; and BghiP, benzo[ghi]perylene.

### 3.3. Human Exposure to Plasticizers and PAHs in Resuspended Dust

Ingestion is the main pathway for intake of plasticizers from dust (Table 4). Regardless of the route, household residents are exposed to higher intakes in the bedrooms, whereas the lowest doses are experienced in the kitchens. The daily intakes for children and adults (13 to 29 and 1.4 to 3.1  $\mu\text{g kg}^{-1} \text{day}^{-1}$ , respectively) via dust ingestion were three to four orders of magnitude higher than those via inhalation (0.357 to 0.802 and 0.129 to 0.289  $\text{ng kg}^{-1} \text{day}^{-1}$ , respectively) and dermal contact (14.9 to 33.0 and 2.05 to 4.62  $\text{ng kg}^{-1} \text{day}^{-1}$ , respectively). Children are at higher risk of exposure to plasticizers than adults. The total  $\text{DI}_{\text{ing}}$ ,  $\text{DI}_{\text{inh}}$ , and  $\text{DI}_{\text{der}}$  for children were about 9.3, 2.8, and 7.1 times higher than those estimated for adults, respectively. DBP, DNOP, and DEHP contributed the most to daily intakes. The dust ingestion intakes for these compounds were lower than the U.S. EPA maximum acceptable oral doses of 0.1, 0.01, and 0.02  $\text{mg kg}^{-1} \text{day}^{-1}$ , respectively. However, the daily intakes of plasticizers through dust ingestion, inhalation, and dermal contact in this study are higher than those estimated for indoor dust from houses of several Chinese regions [39], and childcare facilities, salons, and homes across the USA [29]. Albar et al. [37] assessed human exposure to phthalates via dust ingestion for the worst-case scenario (with 95<sup>th</sup> percentile levels) for Saudi and Kuwaiti toddlers and adults. The exposure to DEHP, which is cardiotoxic and endocrine disruptor, for Saudi toddlers was estimated to be 37,630  $\text{ng kg}^{-1} \text{day}^{-1}$ , while for Kuwaiti toddlers it was 6722  $\text{ng kg}^{-1} \text{day}^{-1}$ . Similarly, exposure estimates to other PAEs, such as DBP, DIBP, and DNOP, was also higher for Saudi toddlers. In the case of Saudi and Kuwaiti adults, dust ingestion intakes for DEHP were estimated at 1613 and 288  $\text{ng kg}^{-1} \text{day}^{-1}$ , respectively.

**Table 4.** Mean daily intakes ( $\text{ng kg}^{-1} \text{day}^{-1}$ ) of plasticizers via dust ingestion, inhalation, and dermal contact for children and adults.

Room	DMP	DEP	DBP	BBP	DEHA	DEHP	DNOP	DINP	DIDP	$\Sigma$
$\text{DI}_{\text{ing}}\text{-CHILDREN}$										
Kitchens	1.78	934	3695	41.4	112	1782	6095	67.1	43.4	12,772
Bedrooms	2.17	296	15,472	68.4	237	5898	6372	131	232	28,707
Living rooms	6.12	914	7765	73.0	125	8890	4938	185	437	23,333
$\text{DI}_{\text{inh}}\text{-CHILDREN}$										
Kitchens	0.000	0.0261	0.103	0.003	0.001	0.050	0.170	0.002	0.001	0.357
Bedrooms	0.000	0.0083	0.432	0.007	0.002	0.165	0.178	0.004	0.006	0.802
Living rooms	0.000	0.0255	0.217	0.003	0.002	0.248	0.138	0.005	0.012	0.652
$\text{DI}_{\text{der}}\text{-CHILDREN}$										
Kitchens	0.002	1.07	4.25	0.129	0.048	2.05	7.01	0.077	0.050	14.7
Bedrooms	0.003	0.340	17.8	0.272	0.079	6.78	7.33	0.150	0.267	33.0
Living rooms	0.007	1.05	8.93	0.144	0.084	10.2	5.68	0.213	0.502	26.8
$\text{DI}_{\text{ing}}\text{-ADULTS}$										
Kitchens	0.190	100	396	12.0	4.44	191	653	7.19	4.65	1368
Bedrooms	0.232	31.7	1658	25.4	7.33	632	683	14.0	24.9	3076
Living rooms	0.655	97.9	832	13.4	7.82	952	529	19.9	46.8	2500
$\text{DI}_{\text{inh}}\text{-ADULTS}$										
Kitchens	0.000	0.009	0.037	0.001	0.000	0.018	0.062	0.001	0.000	0.129
Bedrooms	0.000	0.003	0.156	0.002	0.001	0.059	0.064	0.001	0.002	0.289
Living rooms	0.000	0.009	0.078	0.001	0.001	0.090	0.050	0.002	0.004	0.235
$\text{DI}_{\text{der}}\text{-ADULTS}$										
Kitchens	0.000	0.150	0.595	0.018	0.0067	0.287	0.981	0.011	0.007	2.05
Bedrooms	0.000	0.048	2.49	0.038	0.0110	0.949	1.03	0.021	0.037	4.62
Living rooms	0.001	0.147	1.25	0.020	0.0117	1.43	0.794	0.030	0.070	3.75

Reference doses for oral exposure,  $\text{RfD}$  ( $\text{mg kg}^{-1} \text{day}^{-1}$ ) recommended by the United States Environmental Protection Agency: DEP, 0.8; DBP, 0.1; BBP, 0.2; DEHA, 0.6; DEHP, 0.02; and DNOP, 0.01. Reference doses for other compounds or other exposure pathways are not available.

BaP, DahA, and IcdP were the compounds that most contributed to the carcinogenic potency, accounting for 45% to 57%, 11% to 29% and 11% to 13% of the total BaP<sub>TEQ</sub>, respectively. As shown in Table 5, the total cancer risk could be attributed almost entirely to ingestion and did not vary much with the microenvironment. Therefore, inhalation of resuspended particles through the mouth and nose or via dermal contact was almost negligible as compared with the ingestion route. Under most regulatory programs, an ILCR between  $10^{-6}$  and  $10^{-4}$  indicates potential risk, an ILCR of  $10^{-6}$  or less is considered insignificant, and an ILCR  $\geq 10^{-4}$  is taken as high risk [93,94]. In the present study, the total risk of adult and children exposure to PAHs in dust via the three pathways ranged from  $7.2 \times 10^{-6}$  to  $1.4 \times 10^{-5}$ . This means that the resuspendable thoracic fraction of household dust can contribute to an estimated excess of 7.2 to 14 per million people new cancer cases. One cancer case per million people is usually used as a baseline level of acceptable risk.

**Table 5.** Incremental lifetime cancer risk from human exposure to PAHs in PM<sub>10</sub> resuspended from household dust via ingestion, inhalation, and dermal absorption.

Cancer risk		Kitchens	Bedrooms	Living Rooms
ILCR <sub>ing</sub>	Children	$9.2 \times 10^{-6}$	$7.2 \times 10^{-6}$	$9.4 \times 10^{-6}$
	Adults	$1.4 \times 10^{-5}$	$1.1 \times 10^{-5}$	$1.4 \times 10^{-5}$
ILCR <sub>inh</sub>	Children	$1.4 \times 10^{-10}$	$1.1 \times 10^{-10}$	$1.4 \times 10^{-10}$
	Adults	$6.9 \times 10^{-10}$	$5.4 \times 10^{-10}$	$9.0 \times 10^{-10}$
ILCR <sub>der</sub>	Children	$3.6 \times 10^{-8}$	$2.8 \times 10^{-8}$	$3.7 \times 10^{-8}$
	Adults	$7.1 \times 10^{-8}$	$5.6 \times 10^{-8}$	$7.3 \times 10^{-8}$
Total ILCR	Children	$9.3 \times 10^{-6}$	$7.2 \times 10^{-6}$	$9.4 \times 10^{-6}$
	Adults	$1.4 \times 10^{-5}$	$1.1 \times 10^{-5}$	$1.4 \times 10^{-5}$

To assess the potential health risk of foliar dust in Nanjing, China, Zha et al. [56] analyzed the contents of 16 priority PAHs. Total concentrations in dust ranged from 1.77 to 19.02  $\mu\text{g g}^{-1}$ , with an average value of 6.98  $\mu\text{g g}^{-1}$ . The cancer risk levels via dermal contact and ingestion varied from  $10^{-8}$  to  $10^{-6}$  in all the dust samples, while the mean cancer risk via inhalation was  $10^{-10}$  to  $10^{-12}$ , about  $10^4$  to  $10^7$  times lower than through ingestion and dermal contact.

ILCR values due to human exposure to PAHs in indoor dust in city, town, village, and orefield of Guizhou province, China, were  $6.14 \times 10^{-6}$ ,  $5.00 \times 10^{-6}$ ,  $3.08 \times 10^{-6}$ , and  $6.02 \times 10^{-6}$  for children and  $5.92 \times 10^{-6}$ ,  $4.83 \times 10^{-6}$ ,  $2.97 \times 10^{-6}$ , and  $5.81 \times 10^{-6}$  for adults, respectively [16]. As noted in our study, inhalation of resuspended particles through mouth and nose was very small as compared with the ingestion pathway. Maertens et al. [87] estimated higher excess lifetime cancer risks, ranging from  $1 \times 10^{-6}$  and  $1 \times 10^{-4}$ , associated with nondietary ingestion of PAHs in settled dust from homes in Ottawa, Canada, during preschool years. However, the researchers observed that the level of risk varies substantially according to the ingestion rates adopted to perform the estimates.

Since ingestion was found to be the dominant exposure pathway, the risk associated with seven non-carcinogenic PAHs (ACY, ACE, FLU, PHE, ANT, FLUA, and PYR) in household PM<sub>10</sub> dust was estimated through the hazard quotient (HQ). Under most programs, if the HQ value is greater than one, the exposed population is likely to experience considerable non-carcinogenic effects. The highest daily intakes were obtained for PYR (24 to 31  $\text{ng kg}^{-1} \text{day}^{-1}$  for children and 2.6 to 3.3  $\text{ng kg}^{-1} \text{day}^{-1}$  for adults) and ACY (12 to 31  $\text{ng kg}^{-1} \text{day}^{-1}$  for children and 1.2 to 3.3  $\text{ng kg}^{-1} \text{day}^{-1}$  for adults). These values are higher than those documented for dust samples collected from three different microenvironments (cars, air conditioner filters, and household floor dust) of Jeddah, Saudi Arabia, and Kuwait [92]. Nevertheless, the HQ values of our study were less than one, indicating that there was no considerable non-carcinogenic risk arising from ingestion of PAHs in resuspended PM<sub>10</sub>. Although the risk remains very low for both age groups, higher HQ values for children ( $1.51 \times 10^{-3}$  to  $1.89 \times 10^{-3}$ ) than those

obtained for adults ( $1.32 \times 10^{-4}$  to  $2.03 \times 10^{-4}$ ), demonstrate greater susceptibility of the younger ones. It should be noted that HQ values are underestimated as naphthalene was not included in the quantifications. Despite its volatility, naphthalene is normally quite abundant in the particulate phase.

#### 4. Conclusions

This preliminary study provides a first insight on the occurrence of plasticizers and PAHs in PM<sub>10</sub> from resuspended dust samples in Spanish households and adds to the growing evidence that non-dietary exposure contributes to the total body burden. Considering that people spend most of their time indoors, exposure to these pollutants could lead to an increased human health risk. Although no appreciable differences between plasticizer and PAH levels in resuspended dust from the various residential microenvironments were observed, it was concluded that exposure through the ingestion route poses much higher risks as compared to inhalation and dermal contact. This is of particular concern for infants due to their higher dust intake via frequent hand-to-mouth activities. Because of the small number of samples analyzed in this study, it should be noted that exposure estimates are only an indication of the likely range for children and adults within the studied population. More assessments with wider spatial and temporal coverage are needed to better understand the dynamics and possible effects of these pollutants in different indoor microenvironments. As observed in other works, this study on exposure to organic pollutants suggests that measurements only in food and outdoor environments can substantially underestimate exposures to chemicals. Comparisons of the results of this study with those reported in the literature revealed huge amplitudes in the numbers and great difficulty in generalizing conclusions, mainly due to the heterogeneity of applied methodologies. Traditional methodologies have been compartmentalized (settled versus suspended dust) and as a result may have described the environment incompletely. Thus, the scientific community and international agencies should discuss and establish standardized protocols.

**Supplementary Materials:** The following are available online at <http://www.mdpi.com/2073-4433/10/12/785/s1>, Analytical technique for quantification of the carbonaceous content of PM<sub>10</sub>, Table S1: Standards from Sigma-Aldrich (and product references) used in chromatographic analysis, Table S2: Dust loading obtained in the various houses, in both sampling periods, mass percentage of carbonaceous material (TC = OC + EC) and mass fractions of plasticizers and polycyclic aromatic hydrocarbons in PM<sub>10</sub>.

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