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Polycyclic aromatic hydrocarbons, polychlorinated biphenyls and legacy and current pesticides in indoor environment in Australia – occurrence, sources and exposure risks

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13 **Highlights**

14 A modified PAS was used to simultaneously collect gaseous SVOCs and particles;

15 PCB 11 was identified and quantified in indoor environments for the first time;

16 PCBs and pesticides were from indoor while PAHs were influenced by outdoor sources;

17 Indoor pesticide levels in Australia were among the highest in the world;

18 The highest EDI value was estimated for permethrin, mostly via exposure to dust.
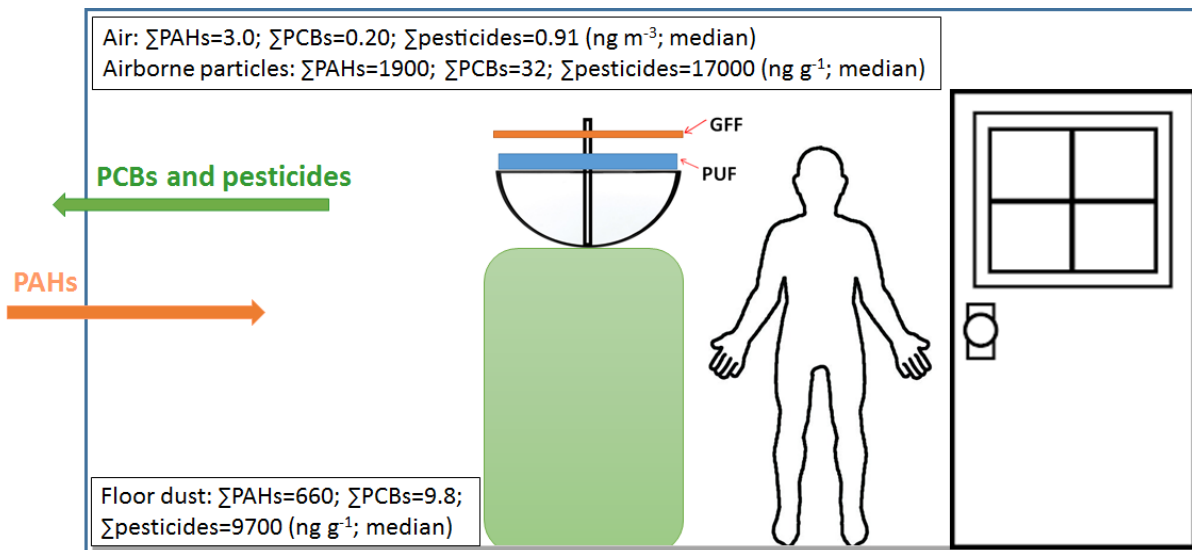
19   **Abstract**

20   Paired indoor air and floor dust samples were collected from residential houses and offices (n = 28) in

21   two Australian cities in 2015. For the air samples, a modified passive air sampler (PAS) was used to

22   collect semi-volatile organic compounds (SVOCs) in gaseous phase and airborne particles

23   simultaneously. Sampling rates (R) of the PAS for gaseous SVOCs ranged from 0.69 to 3.4 m^3

24   sampler^{-1} day^{-1}. Out of the 33 analytes, 22, 14 and 17 compounds were detected (above the method

25   detection limit) in over 50% of air, airborne particles and floor dust samples respectively. The highest

26   median level in air, airborne particles and floor dust was observed for phenanthrene (2.0 ng m^{-3}),

27   permethrin (8800 ng g^{-1}) and permethrin (5100 ng g^{-1}) respectively. Amongst polychlorinated

28   biphenyl (PCB) congeners, with few exceptions, the largest contribution was from 3,3'-

29   dichlorobiphenyl (PCB11) for both indoor air and floor dust samples. In these houses and offices, the

30   indoor level of polycyclic aromatic hydrocarbons (PAHs) was mainly influenced by ambient (outdoor)

31   air. Primary sources of PCBs were from within indoor environments and generally older houses have

32   higher concentrations in air. Amongst pesticides, hexachlorobenzene in indoor environments appeared

33   to be due to transfer from outdoor sources whereas chlordanes and pyrethroids were associated with

34   past and current household application respectively. Compared to data from other countries/regions,

35   concentrations of chlordanes, chlorpyrifos and pyrethroids in indoor air and dust samples from

36   Australia were among the highest whereas PCB and PAH levels were among the lowest. The sum of

37   estimated daily intakes (EDIs) via inhalation and dust contact and ingestion were calculated. The

38   highest median value of EDI was observed for permethrin at 2.8 (for adults) and 74 ng kg^{-1} day^{-1} (for

39   toddlers), which are <0.15 % of the U.S. EPA reference dose.

40   **Keywords**

41   Indoor; semi-volatile organic compounds; passive air sampling; sources; global comparison; daily

42   intakes

Graphical abstract

Air: ∑PAHs=3.0; ∑PCBs=0.20; ∑pesticides=0.91 (ng m^{-3}; median)
Airborne particles: ∑PAHs=1900; ∑PCBs=32; ∑pesticides=17000 (ng g^{-1}; median)

GFF

PUF

PCBs and pesticides

PAHs

Floor dust: ∑PAHs=660; ∑PCBs=9.8; ∑pesticides=9700 (ng g^{-1}; median)

1. Introduction

Indoor environments represent an important source for human exposure to organic chemicals for two reasons: i) people typically spend 70 – 90% of their time within homes and offices (Lucattini et al., 2018; Schweizer et al., 2007); and ii) a wide range of organic chemicals were/are used, and produced in and introduced into indoor environments. Many of these chemicals are semi-volatile organic compounds (SVOCs). Once emitted into indoor environments, these chemicals can disperse among different matrices/surfaces such as air, airborne particles and dust resulting in potential chronic exposure via inhalation, dust ingestion and/or dermal contact (Weschler and Nazaroff, 2008). Indoor environments also typically have less exposure to UV light and thus fewer photolytic reactions, making these chemicals more stable therein (Audy et al., 2018).

Understanding the levels and sources of these chemicals in indoor environments is key for assessing human exposure pathways and risks, and implementing control/intervention processes. Comprehensive data for SVOCs in indoor environments are scarce in Australia and have mostly focused on flame retardants (Harrad et al., 2016; He et al., 2018; Toms et al., 2009). There is a lack of data for many other SVOCs such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and legacy and current pesticides. These chemicals can be introduced into indoor environments from different sources and via multiple pathways. PAHs can be emitted from cooking and smoking, indoor and outdoor biomass burning and vehicular emissions. PCBs and legacy pesticides, although banned for many years, are very persistent and can be emitted into indoor environments from sources such as old but in-use transformers and capacitors as well as building sealant (PCBs) and previously contaminated surfaces in and surrounding the building (OCPs). Current pesticides such as organophosphate pesticides and pyrethroids which are used for pest control in the building may also result in a high level of human exposure in indoor environments. Some compounds in these chemical groups such as benzo(a)pyrene and some PCB congeners are classified as carcinogenic to humans by the International Agency for Research on Cancer (IARC, 2015). Some other chemicals such as pyrethroid pesticides are considered to be endocrine disrupting and linked to various negative health outcomes (Meeker et al., 2008).

Air and dust are two very relevant matrices for investigating the exposure to SVOCs in indoor environments (Weschler and Nazaroff, 2008). Lucattini et al. (2018) conducted a comprehensive review of SVOCs in the indoor environment and presented that concentrations of PAHs, PCBs and pesticides in indoor air and floor dust samples have been reported mostly from countries/regions in the Northern Hemisphere. Knowledge gaps concluded in the above review also included the difficulty in the identification of main sources, as well as a limited (and likely underestimated) data of particle-associated SVOCs. Active air samplers (AAS) can sample both gaseous chemicals and particulates at the same time but their main drawback for indoor sampling, particularly within residential settings, is the noise emitted during operation. Therefore, various PAS have been developed as alternatives to AAS and widely/prevalently used for indoor SVOC sampling since 2004 (Bohlin et al., 2008; Harrad et al., 2006; Wilford et al., 2004) but primarily sample chemicals in the gaseous phase. Reported in a very limited

83   number of studies, effort has been recently made to designing PAS that can sample SVOCs in the particle-
84   associated phase in the air in addition to the gaseous phase to achieve comparable monitoring capability as
85   active air sampling (Abdallah and Harrad, 2010; Tao et al., 2009).

86   In this pilot study, a new modified PAS was used to simultaneously collect gaseous SVOCs and airborne
87   particles from 28 homes and offices in two Australian cities. Paired dust samples were also collected. The
88   current study presents an effort as one of the pioneering studies that systematically and simultaneously
89   monitors multi-class SVOCs in air, particles and dust in indoor environments. To our best knowledge, this
90   is also the first study in Australia monitoring PAHs, PCBs and pesticides in indoor environments from all
91   these three matrices. The aim of this study was to investigate the levels, profiles and distribution of PAHs,
92   PCBs and legacy and current pesticides in indoor air and dust and to evaluate their current major sources
93   and human exposure risks.

94   ## 2. Methodology

95   ### 2.1 Sample collection

96   Paired air and dust samples were collected from 14 residential houses and 14 offices in Brisbane and
97   Canberra, Australia in 2015. Air samples were collected using a modified PAS composed of glass fibre
98   filter (GFF) and polyurethane foam (PUF), respectively (Fig. 1). The GFF mounted on the top of the
99   sampler was held by a metal rack to collect airborne particles and a PUF disk was positioned under the GFF
100 to sample the target chemicals in the gaseous phase as well as potentially the ones associated with fine
101 (with a diameter < 2 μm) particles (Bohlin et al., 2010). Samplers were deployed for 2-3 months at
102 locations away from direct ventilation (i.e. open window and/or air supply outlet of air conditioners) and
103 being mounted at a height of $1.5 - 2$ m above the ground. After collection, GFF and PUF were retrieved and
104 wrapped separately in aluminium foil and then stored at $- 20$ °C until analysis. The study was conducted
105 with the ethics approval from The University of Queensland (2015000153).

106 Collection of floor dust samples were detailed elsewhere (He et al., 2018). Briefly, they were collected
107 using a clean nylon sampling sock with a vacuum cleaner and then sieved using a pre-cleaned 1mm-mesh
108 metal sieve. Typically 0.1 gram of sieved dust was aliquoted from each sample and analysed.
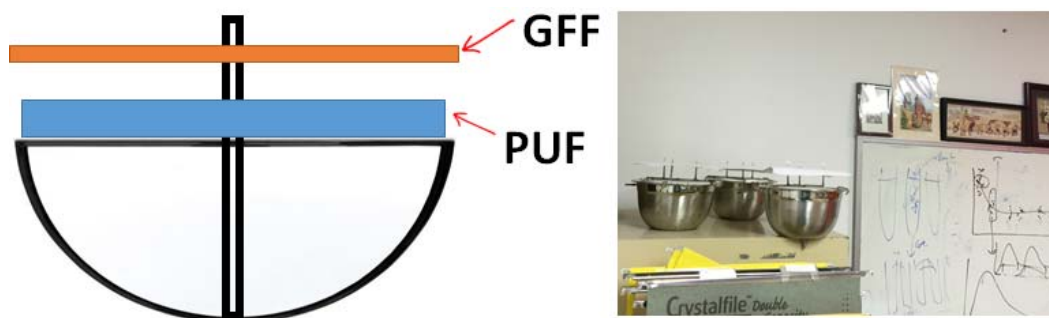
109

110 **Figure 1**. Schematic description and exemplified deployment of the modified PAS used in this study

111 ### 2.2 Chemical analysis

Solvents, reagents and chemical standards were purchased from multiple suppliers. Details are provided in Section S1 of the Supplementary Material (SM). The collected GFF, PUF and dust samples were spiked with 100 μL of an internal standard solution containing multiple deuterated PAHs, ^{13}C-labelled PCB congeners and ^{13}C-labelled pesticides (in isooctane) for quantification purposes (Table S1). Subsequently, these samples were separately extracted via pressurised liquid extraction (PLE) on a Dionex ASE 350 Accelerated Solvent Extractor (Thermo Fisher Scientific) before purified using a chromatographic column. Samples were analysed using a Thermo TRACE GC Ultra coupled to a TSQ Quantum XLS triple quadrupole mass spectrometer equipped with a TriPlus Autosampler. Target analytes include 13 PAH compounds, namely phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo[a]anthrancene+chrysene (BaA+Chr), benzo[b+k]fluoranthene (BbF+BkF), benzo[e]pyrene (BeP), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (I123cdP), dibenzo[a,h]anthracene (DahA), benzo[g,h,i]perylene (BghiP); 8 PCB congeners including PCB 11, 28, 52, 101, 118, 138, 153 and 180; and 12 pesticides including hexachlorobenzene (HCB), heptachlor (HEPT), heptachlor epoxide B (HEPX), *trans*-chlordane (TC), *cis*-chlordane (CC), *p,p'*-DDE, *p,p'*-DDT, chlorpyrifos (CPF), permethrin (PERM), cypermethrin (CYPERM), cyfluthrin (CYF), and deltamethrin (DELTAM). Details of chemical analysis are described in Section S2 in the SM.

2.3 Quality assurance and quality control (QA/QC)

Sampling rate of the PAS. To obtain the sampling rate R (m^3 sampler^{-1} day^{-1}) of the PAS, a low-volume active air sampler (SAICI Technology Co., LTD., LSAM-100; (Wang et al., 2016)) operating at 0.22 m^3 h^{-1} was used to collect air samples in parallel with the PAS deployed at one of the sampling site (Office #1). An XAD cartridge (1 g) was used to trap chemicals in the gaseous phase beneath a GFF collecting particle-associated chemicals. The results from XAD were used to calculate the sampling rate of the PUF for gaseous chemicals. The chemical concentrations in gaseous phase are reported as pg m^{-3} after conversion and the particle-associated chemicals collected by the GFF of the PAS are reported as ng g^{-1} airborne particles.

We assumed a linear-phase sampling condition being maintained for all the chemicals on PAS during the sampling period (50 – 95 days) (Shoeib and Harner, 2002) and calculated the compound-specific sampling rate R from:

$$R = \frac{C_{PAS}/t}{C_{AAS}} \quad (1)$$

where C_{PAS} is the amount of chemicals sequestered by the samplers (in pg sampler^{-1}) during the deployment period t (days). C_{AAS} the volumetric concentrations (in pg m^{-3}) of chemicals derived from the sample collected by the active air sampler. The calculated R was then applied to each sampler deployed on different sampling sites using:

$$C_{Air} = \frac{C_{PAS}}{R \times t} \quad (2)$$

146 where C_{Air} is the converted volumetric concentrations in air (in pg m^{-3}). It should be noted that uncertainties

147 of R existed, due to the differences in ventilation capacity/rate at different homes and offices. Since efforts

148 were made to avoid direct ventilation contact as mentioned above, the impact of wind speed and velocity on

149 R was expected to be minimal.

150 With the active XAD samples, 16 compounds were detected above their respective method detection limit

151 (MDL) and the sampling rate R was derived directly from equation (1). For the other chemicals, R was

152 estimated based on their molecular weights and structures. Detailed estimation method for specific

153 compounds and R value for all the above chemicals (and extended ones) can be found in Table S2. Briefly,

154 R ranged from 0.69 to 3.4 m^3 sampler^{-1} day^{-1} for the compounds targeted in the current study.

155 *Sampling and analytical precision.* Duplicated air samplers were deployed in Office #1 for 94 days and

156 analysed separately to check the precision of sampling and chemical analysis for air samples. Similarly,

157 dust samples from House #1 and Office #20 were respectively homogenised, aliquoted and analysed in

158 duplicate to examine the precision of dust analysis. The precision was estimated based on the relative

159 standard deviation (RSD) between the duplicate samples (i.e. one PUF pair, one GFF pair and two dust

160 pairs). RSD ranged from 0.47% to 53% and for ~90% of the cases it was ⩽30%. Full information is

161 provided in Table S3 in the SM.

162 *Blank samples and method detection limits (MDLs).* Within each batch of samples analysed (typically 8

163 samples per batch), a solvent blank, a matrix blank and a field blank were incorporated to check for any

164 contamination related to analytical instruments, the sample preparation system and transportation and

165 storage of samples. A pre-cleaned PUF, GFF and 0.1 gram of anhydrous sodium sulphate acted as the

166 matrix blank for air, airborne particles and floor dust samples respectively. Non-exposed GFFs and PUFs (n

167 = 6 respectively) that underwent the same procedure of transportation of actual samples were used as the

168 field blank for air samples. Anhydrous sodium sulphate was sealed in an air-tight jar and underwent the

169 same procedure of transportation of actual dust samples to act as the field blank (n = 4) for dust samples.

170 MDLs were defined as the average field blank levels plus three times the standard deviation. If the relevant

171 compounds could not be detected within the field blank samples, MDLs were determined based on half the

172 instrument detection limits (IDLs). MDLs for the analytes ranged from 0.0012 to 140 pg m^{-3} in air, from

173 0.019 to 3400 ng g^{-1} airborne particles and from 0.0020 to 2500 ng g^{-1} floor dust (Table 1).

2.4 Estimation of human intake of SVOCs from inhalation, dust ingestion and dermal contact

175 The equations and parameters used for this estimation were detailed in He et al. (2018) and references

176 therein. That study collected and analysed indoor air and dust samples from a very similar set of sampling

177 locations in the same cities with the current study. Briefly, intake estimation was conducted for toddlers and

178 adults separately, based on a range of parameters/conditions including chemical concentrations (pg m^{-3} air

179 or ng g^{-1} dust), fraction of the day spent at workplace and home (dimensionless), respiration rate (m^3 day^{-1}),

180 body weight (kg), dust ingestion rate (mg day^{-1}), body surface area (cm^2 day^{-1}), soil adhered to skin (mg cm$^-$

181   [2]) and fraction of chemicals absorbed in the skin (dimensionless). Detailed description of these parameters

182   is provided with Table S6 in the SM.

183   **2.5 Statistical analysis**

184   Data analysis was performed using GraphPad Prism 8.0.1. Values lower than the MDL were replaced by ½

185   MDL. Student's *t* test was used to determine differences of concentrations among samples. Criteria for

186   significance was set at $p < 0.05$.

187

188   **3.   Results and discussion**

189   **3.1 SVOCs in indoor air**

190   Overall, out of the 33 analytes measured, 22 and 14 compounds were detected above the MDL in over 50%

191   of gaseous and airborne particle samples respectively (Table 1). Amongst these chemicals, the highest

192   median level in the gaseous phase and airborne particles was observed for phenanthrene (Phe; 2.0 ng m^{-3})

193   and permethrin (PERM; 8800 ng g^{-1}) respectively. Generally, in the same compound group, more volatile

194   compounds were detected with a higher frequency in the gaseous phase and less volatile chemicals showed

195   a higher detection frequency in the airborne particles (Table 1). Gas-particle distribution of target chemicals

196   is discussed in details in Section S3 and Fig. S1 in the SM. In addition, the particle mass (calculated as μg

197   day^{-1}) measured from houses (with a median of 65) was overall higher than the offices (with a median of

198   34). With the outlier (of data from Office #5) excluded from the office dataset, this difference was

199   significant (Mann-Whitney test, $p = 0.014$). This reflected an effectiveness of central ventilation systems in

200   office settings to remove the large particles from the outdoor air being exchanged into indoor environments.

201   It should be noted that our data for airborne particles are expressed as ng g^{-1} particles and thus reflected a

202   particle mass normalised concentration instead of volumetric levels e.g. ng m^{-3}. We recognise this as a

203   limitation of the comparability of our data with other indoor and outdoor studies. This was due to that our

204   sampler design/modification collected "all" the airborne particles instead of size-characterised particles e.g.

205   total suspended particles and there is currently a lack of reliable calibration dataset as we did for the

206   gaseous SVOC sampling and monitoring (Section 2.3). However, with this limitation in mind, the data for

207   SVOCs in airborne particles are still valuable. For example, comparing airborne particle data to dust data

208   (which are reported in the same unit) allows us to estimate SVOC distribution between airborne particles

209   and floor dust, which assists the assessment of contribution from relevant sources (see later sections).

Table 1. Detection frequency (DF), median and mean concentration and method detection limit (MDL) of PCBs, PAHs and pesticides in indoor air (pg m^{-3}), airborne particles (ng g^{-1}) and floor dust (ng g^{-1})

Compounds	Air (pg m^{-3}, n = 28)				Airborne particles (ng g^{-1}, n = 18)				Floor dust (ng g^{-1}, n = 28)			
	DF	Median	Mean ± SD	MDL	DF	Median	Mean ± SD	MDL	DF	Median	Mean ± SD	MDL
Phenanthrene (Phe)	100%	2000	2400 ± 1500	140	11%			3400	Nil			2500
Anthracene (Ant)	93%	190	260 ± 180	20	5.6%			280	Nil			230
Fluoranthene (Flu)	100%	500	640 ± 390	4.3	83%	290	340 ± 210	100	31%			75
Pyrene (Pyr)	100%	400	460 ± 250	9.0	89%	520	700 ± 590	230	62%	140	190 ± 170	45
Benzo[a]anthrancene+chrysene (BaA+Chr)	69%	46	57 ± 29	5.6	89%	280	360 ± 210	91	97%	150	310 ± 410	0.061
Benzo[b+k]fluoranthene (BbF+BkF)	17%			7.7	78%	190	220 ± 120	54	93%	63	120 ± 190	0.092
Benzo[e]pyrene (BeP)	72%	24	95 ± 150	3.1	89%	130	150 ± 80	25	93%	48	480 ± 1500	0.092
Benzo[a]pyrene (BaP)	31%			0.10	44%			0.87	83%	30	54 ± 69	0.092
Indeno[1,2,3-cd]pyrene (I123cdP)	10%			6.3	72%	280	340 ± 200	46	93%	38	76 ± 95	0.38
Dibenzo[a,h]anthracene (DahA)	Nil			0.14	28%			1.3	83%	21	41 ± 50	0.14
Benzo[g,h,i]perylene (BghiP)	69%	35	120 ± 190	2.3	83%	140	200 ± 160	26	93%	79	140 ± 150	0.078
$\sum_{13}$ PAHs	NA	3000	4000 ± 2300		NA	1900	3400 ± 4600		NA	660	1400 ± 2000	
PCB 11	100%	110	150 ± 130	5.2	11%			80	66%	5.6	6.4 ± 2.5	3.2
PCB 28	100%	31	65 ± 85	1.4	17%			20	3.4%			5.4
PCB 52	100%	11	17 ± 14	0.0089	33%			0.17	34%			1.9
PCB 101	97%	11	21 ± 48	0.0064	50%	10	22 ± 22	0.11	28%			1.3
PCB 118	86%	3.5	7.2 ± 6.9	0.22	17%			3.7	69%	1.4	1.7 ± 1.0	0.39
PCB 138	86%	1.5	4.2 ± 7.2	0.0026	22%			0.040	24%			1.3
PCB 153	86%	1.8	6.5 ± 14	0.24	22%			3.7	79%	2.0	2.7 ± 2.0	0.39
PCB 180	45%			0.094	11%			1.3	69%	0.60	1.3 ± 1.7	0.0061
$\sum_{8}$ PCBs	NA	200	270 ± 170		NA	32	60 ± 110		NA	9.8	12 ± 9	

| Compound												
Hexachlorobenzene (HCB)	97%	27	63 ± 98	0.0012	78%	11	42 ± 98	0.019	Nil			0.0020
Heptachlor (HEPT)	93%	45	98 ± 150	0.69	33%			8.6	Nil			530
Heptachlor epoxide B (HEPX)	76%	6.0	26 ± 51	0.52	5.6%			6.2	17%			0.18
Trans-chlordane (TC)	93%	120	210 ± 250	1.1	72%	180	2000 ± 5800	29	3.4%			100
Cis-chlordane (CC)	93%	9.1	31 ± 60	0.063	44%			1.1	3.4%			11
p,p'-DDE	31%			2.7	17%			60	48%			1.4
p,p'-DDT	3.4%			27	28%			610	66%	15	17 ± 16	0.54
Chlorpyrifos (CPF)	90%	260	7100 ± 24000	1.5	78%	150	2800 ± 6300	27	69%	610	2000 ± 3800	120
Permethrin (PERM)	62%	84	260 ± 370	23	100%	8800	19000 ± 39000	420	93%	5100	23000 ± 41000	0.51
Cypermethrin (CYPERM)	3.4%			8.9	11%			160	48%			17
Cyfluthrin (CYF)	6.9%			4.6	44%			85	69%	69	400 ± 1400	8.5
Deltamethrin (DELTAM)	76%	1.1	4.2 ± 5.9	0.079	72%	110	1000 ± 2000	1.4	93%	82	660 ± 1300	0.67
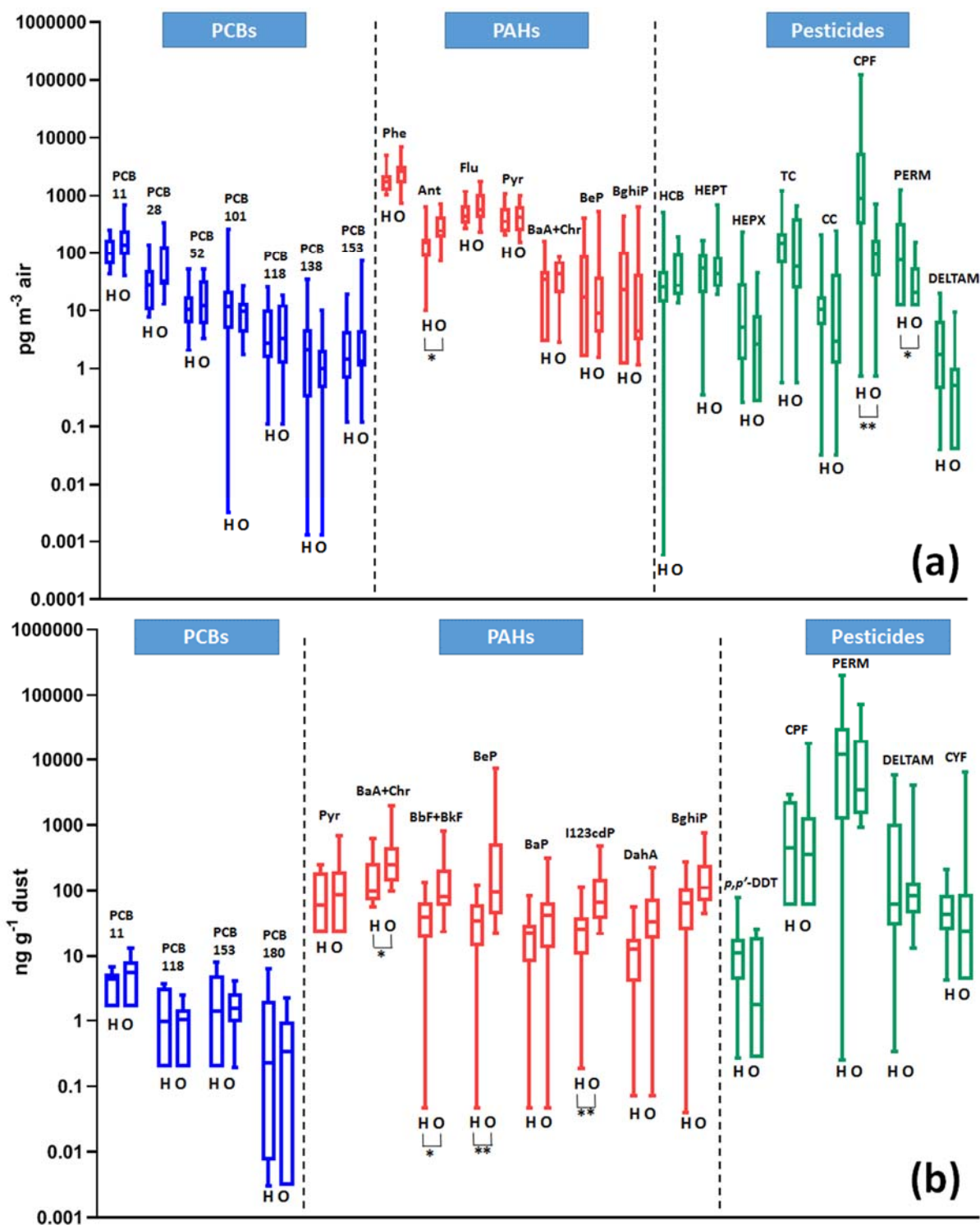
Figure 2. Concentrations presented in log scale in air (Figure 2(a); pg m^{-3}) and floor dust (Figure 2(b); ng g^{-1}) for target compounds in houses (H) and offices (O). Significant difference in concentrations between houses and offices for specific compounds was labelled as * ($p < 0.05$) or ** ($p < 0.01$).

PAHs. In the gaseous phase, concentration of $\sum_{13}$ PAHs ranged from 1.3 to 11 ng m^{-3} with a median level of 3.0 ng m^{-3}. No statistically significant difference can be observed for PAH concentrations (log-transformed) in air between houses and offices with the only exception of anthracene (Ant) whose concentration in the offices was significantly higher than the houses ($p = 0.034$; Fig. 2). A comparison of indoor and outdoor concentration in air was conducted to assist the assessment of sources. We used data

from Wang et al. (2016) who reported the most up-to-date atmospheric PAH and PCB data for Brisbane city.

Generally the concentrations of PAHs measured in these indoor houses and offices were similar to the outdoor levels in Brisbane (with a median value of indoor/outdoor ratio of 0.76 – 1.4 for different compounds; Fig. 3). As previously mentioned, indoor environments are a relatively confined space and potentially have fewer photolytic reactions (Audy et al., 2018), compared to outdoor environments. If any significant PAH source presents in indoor environments, the indoor concentrations of PAHs are expected to be (much) higher than the outdoor environments. For example, wood/fuel burning within residential houses may significantly increase PAH concentrations in homes (Gustafson et al., 2008; Shen et al., 2011), leading to a summed PAH concentration in indoor air at the level of $\mu g/m^3$ (Ding et al., 2012), 3 orders of magnitudes higher than that was observed in the current study. Therefore the indoor/outdoor ratio of 0.76 – 1.4 for PAH concentrations in air may indicate the contribution from generic sources for indoor PAH burdens in air such as exchange from outdoor air, and may also suggest a limited role of cooking and smoking as sources within these indoor settings involved in the current study. A similar observation for PAHs has also been reported by Laborie et al., (2016) and Romagnoli et al., (2014). For offices with controlled ventilation only (i.e. without openable windows), this exchange of PAHs in gaseous phase and that are associated with fine particles from outdoor to indoor may occur through the central filtration system (K. Koponen et al., 2001; Lv and Zhu, 2013; Ohura et al., 2004; Weschler and Nazaroff, 2008). In the airborne particles, concentration of $\sum_{13}$ PAHs ranged from 480 to 20000 ng g^{-1} with a median value of 1900 ng g^{-1}. The highest level of benzo[a]pyrene (BaP) was 100 ng g^{-1}.
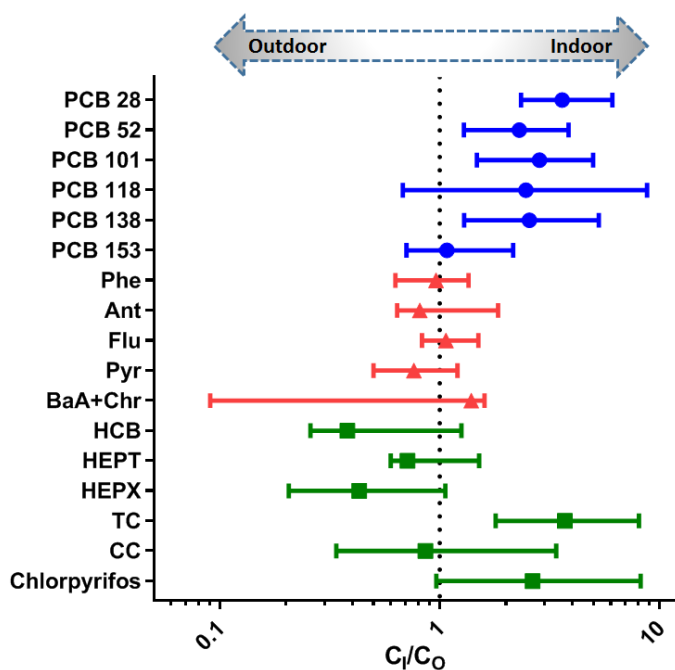
Figure 3. Ratio of concentrations of PCBs (blue dots and lines), PAHs (red triangles and lines) and pesticides (green squares and lines) in indoor air (C_I, current study) and outdoor air (C_O; Wang et al., 2015, 2016 & 2017) in Brisbane. Median values with 95% confidence interval of the ratio for each chemical are shown in logarithmic scale.

PCBs. In the gaseous phase, with a few exceptions, the largest contribution to the sum concentration of PCBs was from 3,3'-dichlorobiphenyl (PCB 11; Fig. 2). To our knowledge, this is the first time that PCB 11 concentrations are measured and reported in indoor environments. PCB 11 is an impurity that is formed during the production of yellow pigments that are contemporarily used in multiple consumer products (Hites, 2018; Hu et al., 2008; Rodenburg et al., 2009; Rodenburg et al., 2010; Shang et al., 2014). This may explain the relatively steady concentration of PCB 11 in ambient air in North America over the last decade, which is in contrast to other congeners such as dioxin-like PCBs (Hites, 2018). In the airborne particle phase, PCB congeners had a low DF (11% – 50%).

PCB data from outdoor air were collated from previous studies (Wang et al., 2015&2016) and compared to their indoor concentrations from the current study. PCB concentrations in indoor air generally exceeded those in outdoor air (with a median value of indoor/outdoor ratio of 1.1 – 3.6 for different congeners; Fig. 3). This may indicate the contribution from indoor emission sources (Australian Government, 2018a; Benthe et al., 1992; Kohler et al., 2005). Similar findings were also reported previously (Li et al., 2018; Marek et al., 2017).

Furthermore, linear regression analysis was applied plotting the age of building (converted to number of Julian days at 1st Jan of the year built) and concentrations of PCBs and legacy pesticides in indoor air. Ten houses with relatively high DF of these chemicals were included with ages ranging from 12 to 50 years. Overall, an increase in PCB concentrations was observed with older age (Fig. S2), suggesting that older houses have higher PCB concentrations. It should be noted that, however, no statistical significance could be observed (p = 0.089 – 0.82).

PCB concentrations in indoor air from the current study (Table S4) were at the lower end when compared to other studies in Europe, and North and Central America (Audy et al., 2018; Bohlin et al., 2008), reflecting the historical usage and emissions of PCBs being generally less in the Southern compared to the Northern Hemisphere (Breivik et al., 2002). Similar findings were also reported for outdoor ambient air (Wang et al., 2015).

Pesticides. In the gaseous phase, various legacy pesticides that have been banned in Australia for 2 – 4 decades including hexachlorobenzene (HCB), heptachlor (HEPT) and chlordanes were detected in >75% of samples. The highest median concentration was measured for *trans*-chlordane (TC) at 120 pg m^{-3} (Fig. 2). This value was at the higher end compared to data for Europe and Central America (Table S4). This was in line with the finding from outdoor air (Wang et al., 2015) and might be a reflection of the relatively higher usage of this pesticide historically in Australia. For the current pesticides, chlorpyrifos (CPF) had a relatively high concentration ranging from 25 to 120000 pg m^{-3} with a median level of 260 pg m^{-3} (Fig. 2).

In the airborne particle phase, legacy pesticides generally had a lower DF but the highest median level was still measured for TC (180 ng g^{-1}). The current pesticide PERM was detected in all the samples with a much higher median level of 8800 ng g^{-1} mirroring its current use for domestic pest control.

Concentrations of gaseous pesticides were consistently higher in houses than in offices, with a statistical significance observed for CPF and PERM (p = 0.0021 and 0.023 respectively for log transformed data; Fig. 2). This may again reflect their use pattern in different environments.

Trans- and *cis*-chlordane (TC and CC) are components contained in technical chlordane mixtures in a ratio of approximately 1.2 (Bidleman et al., 2002). Technical chlordane used to be applied for termite control (Kookana et al., 1998; Radcliffe, 2002) in Australia with registration being cancelled in 1997 (Australian Government, 2018b). The much higher ratio of TC/CC in the gaseous phase in this study (1.8 – 210) was an indication of close vicinity to source areas because TC has a higher vapour pressure than CC (Shen and Wania, 2005). In addition, this range was much wider (and overall higher) than the one for outdoor air across Australia (0.97 – 4.9; (Wang et al., 2015)). These results suggested that the major source of chlordanes in indoor environments in Australia maybe the volatilisation from previously treated materials (Shunthirasingham et al., 2010) within/around houses and offices instead of being from outdoor pathways (e.g. long-range atmospheric transport (LRAT)). In addition, the highest value of C_I/C_O was observed for TC in the pesticide group, which provides a further evidence of indoor-environment associated sources being dominant for contributing to its burdens in indoor air. The currently used pesticide CPF had a similar indoor-outdoor concentration ratio range (Fig. 3), reflecting also predominant sources in indoor settings, particularly the residential houses as discussed above. Wang et al. (2017) also attempted measuring outdoor concentrations of PERM in Brisbane and the values were all below 2.1 pg m^{-3} whereas in this current study it had a median concentration at 84 pg m^{-3}. This indicated the same indoor source dominance scenario for PERM as for CPF. On the other hand, the lowest ratio of C_I/C_O was found for HCB, which may mean a prominent contribution from outdoor air to indoor environments.

As with PCBs, a decreasing concentration of legacy pesticides was observed with newer houses but none was statistically significant (p = 0.16 – 0.59).

3.2 SVOCs in floor dust

Overall, 17 out of 33 compounds were detected above the MDL with a DF of > 50%. PERM had the highest median concentration (5100 ng g^{-1}) followed by CPF (610 ng g^{-1}) as shown in Fig. 2.

PAHs. Heavier PAH compounds were frequently detected in the floor dust samples. The median of BaP concentration was 30 ng g^{-1}, less than half of the value reported in dust samples collected 10 years ago from houses in the same city (Robertson et al., 2005) (Table S5). Concentrations of PAH compounds in floor dust from offices were consistently higher than houses. Amongst these, benzo[a]anthrancene+chrysene (BaA+Chr), benzo[b+k]fluoranthene (BbF+BkF), benzo[e]pyrene (BeP) and indeno[1,2,3-cd]pyrene (I123cdP) were significantly higher (in log-transformed concentrations; p = 0.0070 – 0.020) in offices than houses (Fig. 2). In addition, the ratio of $C_{BaP}/(C_{BaP} + C_{BeP})$ was significantly higher in offices (0.41 ± 0.02) than houses (0.37 ± 0.02) (p = 0.049), indicating more contribution from relatively fresher particle emissions (Oliveira et al., 2011) in the offices. These may suggest that offices were less prone to the

influence of outdoor PAHs than homes (Romagnoli et al., 2014) and emission sources for PAHs within the offices such as operation of office equipment (Destaillats et al., 2008) may contribute to their concentrations in dust. No significant correlation of $C_{floor\,dust}$ and C_{air} was observed for any PAH compound, which indicated a non-equilibrium distribution of these chemicals between air and floor dust (Harrad et al., 2009; Tao et al., 2016). A detailed discussion is provided in Section S3 in the SM. No significant correlation was observed between $C_{floor\,dust}$ and $C_{airborne\,dust}$ for any target PAH, indicating a non-equilibrium distribution for these chemicals between airborne particles and floor dust and/or different source patterns. In 90% of cases the ratio of $C_{airborne\,dust}/C_{floor\,dust}$ was equal to or higher than 1. This result reinforced the previously discussed theory that indoor PAH burdens may mainly come from outdoor contribution from ambient air. Overall the levels of PAHs measured from the current study were 10 – 100 times lower than dust in Nigeria and China but 40-50 times higher than Czech Republic (Table S4) (Iwegbue et al., 2018; Melymuk et al., 2016; Wang et al., 2013).

PCBs. Similar to indoor air, PCB 11 still had the highest contribution towards total PCB content with a median value of 57% but was below the MDL for a few sites. No significant difference in concentration was observed for any PCB congener between houses and offices (Fig. 2). PCB concentrations from the current study were at the lower end of the range when compared to other studies in Europe and China (Table S4). Again, no significant correlation of $C_{floor\,dust}$ and C_{air} was observed for any PCB congener, suggesting on-going emissions in the indoor environment.

Pesticides. The only legacy pesticide that had a DF of >50% was *p,p'*-DDT with a median concentration at 15 ng g^{-1}. In contrast, CPF and pyrethroids such as PERM, deltamethrin (DELTAM) and cyfluthrin (CYF) had a higher DF and 4.5 – 330 times higher median concentration. This reflected the use pattern of these pesticides, i.e. that DDT has been banned for general use in Australia since 1987 (Radcliffe, 2002) whereas CPF, PERM, DELTAM and CYF are all currently used pesticides (APVMA, 2018). Pesticide concentrations in dust samples were not significantly different between houses and offices (Fig. 2). It should be noted, however, that the DF of cypermethrin (CYPERM) was approximately 70% in houses, compared to approximately 30% in offices. This may indicate the major use of this pyrethroid in domestic application. Unlike PAHs and PCBs, significant correlations were observed for log-transformed concentrations of various current pesticides including CPF (*p* = 0.021), PERM (*p* = 0.0063) and DELTAM (*p* = 0.00080) between $C_{floor\,dust}$ and C_{air}. This might be a reflection of the indoor use of these pesticides (e.g. for pest control) and relatively faster equilibrium post application between different phases due to their relatively low persistence. On the other hand, a significant correlation (*p* = 0.019) was observed for concentrations of DELTAM between $C_{floor\,dust}$ and $C_{airborne\,dust}$, suggesting an equilibrium condition for this pesticide. Compared to available data from Europe and USA, levels of pyrethroids and CPF from the current study were higher (Table S4), indicating a relatively higher usage of these pesticides in Australia.

3.3 Human exposure estimation

Generally, inhalation is a more important pathway of exposure to more volatile compounds for both adults and toddlers (Table 2). For PCB congeners, the highest estimated daily intakes (EDIs) were calculated for PCB 11 for both adults (30 pg kg (b.w.)$^{-1}$ day^{-1}) and toddlers (57 pg kg (b.w.)$^{-1}$ day^{-1}), mostly via inhalation. Pyr had the highest EDI amongst PAHs, namely 180 and 880 pg kg (b.w.)$^{-1}$ day^{-1} for adults (mostly via inhalation and dust contact) and toddlers (mostly via dust contact and ingestion), respectively.

For currently used pesticides including CPF and pyrethroids (e.g. PERM and DELTAM), dust contact and ingestion contributed > 80% to the sum EDIs for both adults and toddlers (Table 2). This reflected their main use patterns (e.g. as active ingredients in pest control products being sprayed onto floor and lower wall surfaces) and after which their affinity with indoor dust. Toddlers overall had relatively higher EDIs than adults via all routes. The highest ratio of toddler EDI against adult EDI was identified for PERM at 27, mostly through dust contact and ingestion. Overall, the sum of EDIs via inhalation, dust contact and ingestion from indoor environments from the current study were 3 – 5 orders of magnitudes lower than reference dose (RfD; oral), indicating a negligible exposure risk.

Table 2. Exposure risk estimation for adults and toddlers via inhalation, dermal contact with dust and ingestion of dust from indoor environments (median values in pg kg (b.w.)$^{-1}$ day^{-1} for chemicals with a >50% DF only). Values at 5% and 95% were also calculated and shown in Table S6 in the SM.

	Inhalation		Dust contact		Dust ingestion		Sum		RfD (oral) (USEPA, 2018)
	Adult	Toddler	Adult	Toddler	Adult	Toddler	Adult	Toddler	
PCB 11	26	31	4.0	11	0.18	15	30	57	NA
PCB 28	7.1	8.9	NA	NA	NA	NA	NA	NA	NA
PCB 52	2.6	3.4	NA	NA	NA	NA	NA	NA	NA
PCB 101	2.4	4.2	NA	NA	NA	NA	NA	NA	NA
PCB 118	0.80	0.97	0.99	4.8	0.044	6.4	1.8	12	NA
PCB 153	0.41	0.84	1.4	6.9	0.064	9.2	1.9	17	NA
PCB 138	0.35	0.98	NA	NA	NA	NA	NA	NA	NA
PCB 180	NA	0.16	0.43	1.2	0.019	1.6	NA	3.0	NA
$\sum_8$ PCBs	40	51	6.8	24	0.31	32	47	110	$4.5\times10^{4*}$
Phe	450	570	NA	NA	NA	NA	NA	NA	NA
Ant	43	52	NA	NA	NA	NA	NA	NA	3×10^8
Flu	120	140	NA	NA	NA	NA	NA	NA	4×10^7
Pyr	91	120	89	310	4.3	450	180	880	3×10^7
BaA+Chr	11	15	99	230	4.8	330	110	580	NA
BbF+BkF	NA	NA	42	93	2.0	130	NA	NA	NA
BeP	5.4	12	32	80	1.5	110	39	200	NA
BaP	NA	NA	20	52	0.95	74	NA	NA	3×10^5
I123cdP	NA	NA	25	60	1.2	85	NA	NA	NA
DahA	NA	NA	14	32	0.66	45	NA	NA	NA
BghiP	8.0	16	52	150	2.5	220	63	390	NA
$\sum_{13}$ PAHs	730	930	370	1000	18	1400	1100	3300	NA
HCB	6.2	8.9	NA	NA	NA	NA	NA	NA	8×10^5
HEPT	10	24	NA	NA	NA	NA	NA	NA	5×10^5
HEPX	1.4	2.9	NA	NA	NA	NA	NA	NA	1.3×10^4
TC	27	48	NA	NA	NA	NA	NA	NA	5×10^5
CC	2.1	3.6	NA	NA	NA	NA	NA	NA	5×10^5
p,p'-DDE	NA	NA	NA	1.4	NA	8.5	NA	NA	NA
p,p'-DDT	NA	NA	2.3	6.6	0.48	41	NA	NA	5×10^5
CPF	60	310	310	1400	20	2500	390	4200	NA[#]
PERM	19	78	2600	26000	160	48000	2800	74000	5×10^7

CYPERM	NA	NA	NA	470	NA	880	NA	NA	NA
CYF	NA	NA	35	96	2.2	180	NA	NA	NA
DELTAM	0.26	0.71	41	140	2.6	270	44	410	NA

[*] Averaged from the values for Arochlor 1016 and 1254; [#] RfD value for chlorpyrifos was estimated at $10×10^9$ but removed from the assessment system in 2011

4. Conclusions

The chemical cocktail of indoor environments consists of both legacy and emerging organic contaminants, seeing a ubiquitous detection of PCBs, PAHs and pesticides in Australian houses and offices. Their sources, however, are different – PCBs and some legacy (e.g. chlordane) and most of current pesticides are mainly from indoor sources while PAHs and some other legacy pesticides (e.g. HCB) are mainly from outdoor transportation. Although compared to other countries/regions, chlordane, chlorpyrifos and pyrethoid levels in Australian indoor environments are among the highest, human exposure risk remains negligible. Future studies are warranted to further improve the design of PAS collecting size-characterised particles and/or presenting calibration datasets for normalising airborne particle data to volumetric values e.g. ng m^{-3} to improve the comparability with other indoor and outdoor studies. Overall, simultaneous collection and analysis of gaseous phase, airborne particles and floor dust are an effective approach to estimate indoor burdens of SVOCs and evaluate potential sources.

Acknowledgements

Appendix A. Supplementary Material

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