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PCB in air, dust and surface wipes in 73 Danish homes

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As part of an investigation of the total indoor exposure of residents in PCB contaminated apartments in a building estate in Denmark, 73 homes were visited. Samples of air, vacuum cleaner dust and surface wipes were taken and analysed for 15 PCB congeners. The 53 apartments located in high-rise buildings with indoor PCB containing sealants showed significant elevated total concentrations of PCB in air, dust and surface wipes compared to 20 reference homes. The average concentration in exposed homes was 2330 ng PCB_{total}/m^3 in air, 12.000 ng PCB_{sum15}/g in dust and for surface wipes 529 ng PCB_{sum15}/wipe. These values were respectively 40, 25 and 50 times higher than the averages found in reference homes. The air concentration in all exposed homes was higher than the lower recommended action level of 300 ng PCB_{total}/m^3 from the Danish Health Authority, whereas all reference homes were below. The air concentrations in exposed homes were variating, partly due to indoor temperature. The concentrations in air and dust samples were significantly correlated for PCB_{sum7} in exposed homes. No significant correlation was observed between samples of air and surface wipes, despite surface wipes from exposed homes were significantly different from wipes taken in reference homes. Both dust samples and surface wipes have a potential as screening tool for identifying elevated levels of the less chlorinated PCBs in air. Additional short-term measurements of air concentrations in a subgroup of exposed homes indicated limited diurnal variations within the individual homes. The congener patterns were rather uniform within the respective media from the exposed homes and dominated by tri- and tetraPCBs. The pattern of air differed from that of dust and surface wipes, which were more alike. The study demonstrates a wide spread PCB contamination of the indoor environment in buildings with PCB in indoor sealants and confirms the need for identifying and alleviate PCB in buildings causing a health concern to the occupants.

Abstract

1. Introduction

Polychlorinated biphenyls (PCBs) are persistent organic pollutants (POP) and high persistence together with bioaccumulation have made PCB ubiquitous and a global environmental problem. PCBs were produced commercially from 1929 and from the 1950s the production increased drastically, though after the discovery of their widespread environmental impact in the 1970s the production decreased and ended in 1993 (Tanabe, 1988; Breivik et al., 2002). The Stockholm Convention on Persistent Environmental Pollutants has PCB among the initial 12 POPs recognized as causing adverse effects on humans and ecosystems. Most countries have joined the convention, which came into force in 2004 and prohibits the production of PCB and regulates how to handle and dispose of PCB-containing waste. In Denmark, "open use" of PCB in sealants and other construction products was banned in 1977, while all uses including enclosures in transformers and capacitors, were banned in 1986 (Danish Ministry of Environment, 1976, 1986).

PCB had numerous commercial applications, including addition to polymeric building materials as plasticizer or flame retardant. Further, PCBs have found use as a dielectric fluid in capacitors in fluorescent lighting ballasts (Breivik et al., 2002). These uses have shown to have the potential to substantially contaminate the indoor air with PCB (Balfanz et al., 1993; Kohler et al., 2005; Frederiksen et al., 2012; Grontmij & COWI, 2013). With PCBs belonging to the group of Semi Volatile Organic Compounds (SVOC), the presence in air leads to a widespread contamination of the building due to redistribution from the original source to all interior surfaces. The physical-chemical properties enable these compounds to evaporate and subsequently adsorb and desorb from all kinds of surfaces in the indoor

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environment, including paint, surface dust and particles in the air (Weschler and Nazaroff, 2008, 2010). The contamination of interior surfaces from the air is often described as "tertiary" sources to PCB, while PCB migrating from an original source to adjacent material is described as "secondary" sources. Remediation can be a challenge as removal of original sources is often inadequate to substantial lower indoor contamination of the air (Guo, 2012; Grontmij & COWI, 2013; Kolarik et al., 2014; Lyng et al., 2015).

In Denmark the Health Authority considers long-term exposure of air concentrations in the interval 300–3000 ng/m^3 as harmful to health and action to reduce concentration is advised. Concentrations above 3000 ng/m^3 are considered to be an increased health risk and immediate action to reduce concentration is advised (Danish Health and Medicines Authority, 2013). The air concentration of PCB in Denmark and many other European countries is often described as "PCB_{total}", defined as PCB_{sum6} or PCB_{sum7} times a factor of 5 (Verein Deutscher Ingenieure, 2009). PCB_{sum6} is the sum of the congeners PCB-28, -52, −101, −138, −153 and −180, whereas PCB_{sum7} also includes PCB-118 having dioxin like properties (Heinzow et al., 2007). For the lower chlorinated PCB products, the content of PCB-118 is low relative to PCB_{sum6} and therefore, PCB_{sum6} and PCB_{sum7} is very similar (Takasuga et al., 2006). Estimating the total amount of PCB based on PCB_{sum7}, the factor "5" is used to compensate for all the congeners not quantified. In Denmark, PCB contamination in the indoor environment has been a subject of great concern within the last 10 years. Taking all buildings in Denmark into consideration, it is estimated that 0.6–1.7% has air concentration levels exceeding the lower recommended action level (Grontmij and COWI, 2013). Among other things, one great challenge is to identify the buildings. Often buildings are examined by chemical analysis of building materials or by air samples done by professionals. A more simplified screening tool could ease this process. Another challenge is to determine the long-term air concentration levels. Many of the initial air measurements done to identify potential elevated levels are taken as short-term (often 4-h) determinations. Therefore, we also have an interest in more knowledge about variations, diurnal as well as seasonal, in order to evaluate the representability of the air samples.

Recently, almost 40 years after the Danish ban of using PCB in construction products, Meyer et al. (2013) documented that people living in buildings with substantial PCB levels in building materials are getting the majority of their total PCB-burden from exposures in their home. For some congeners, the blood levels were 100 times higher than observed in the reference group. However, little is known about the importance of different exposure routes and parameters of importance for exposure in the home. In order to investigate this, a survey with samples of blood and hand wipes from residents in contaminated apartments have been conducted in the building estate "Brøndby Strand Parkerne" in Denmark. Further, samples of air, vacuum cleaner dust and surface wipes have been collected from their homes. People living in the area in similar surroundings, though without elevated PCB in indoor air, were participating as reference group. The samples from the homes have been analysed for a range of PCB congeners with relevance for indoor air contamination and the purpose of the present paper is to present the results together with an evaluation of dust and surface wipes as screening tools for elevated concentrations of the less chlorinated PCBs in air.

2. Materials and method

2.1. The buildings

The study was conducted in a building estate with 15-story apartment buildings in Brøndby Strand Parkerne in Denmark, erected in 1969–1974. The building estate consists of 12 similar high-rise buildings, with identical floorplans comprising four apartments in sizes 64, 77, 103 and 129 m^2. A few of the 129 m^2 apartments have been divided into two apartments. The construction principle is load-bearing partitioning walls and façades comprising both sandwich concrete elements and light façade structures. Windows are mounted both in concrete elements and in the light façade structures. The five buildings erected first have PCB in sealants around the light façade elements indoors and outside on the glazed balcony and some windows (Brøndby Strand Parkerne, Brøndby Boligselskab, 2013; Golder Associates A/S, 2017). The buildings have central heating and the pipes are located below the wooden floors, where also some of the sealants are situated (see SM, Fig. S1). The seven subsequently erected buildings are without PCB as additive in the sealants. After awareness of substantial contamination of the indoor air in the five buildings with PCB containing sealants, a pilot remediation has been conducted in two abandoned apartments. During this process, old spillage of PCB on the concrete slabs below the wooden floors was observed as an additional source. The "reference homes" in the investigation were located in the high-rise buildings without PCB containing sealant as well as in surrounding lower buildings and one single family detached house, all erected in the same time period, though without PCB in the building materials.

2.2. Home visits

The survey was conducted in the cold season with central heating on as this was expected to give the least variation in indoor room temperature during the period of air sampling. Altogether 73 homes were visited for sampling and temperature measurements, 53 exposed and 20 reference homes. One exposed apartment was in use, but not for accommodation, though as the results looked similar to other homes it was included in the study. Participants were asked to behave as usual during the period of air sampling. They were also asked to fill in a questionnaire about airing and cleaning behaviour. The homes were visited on two consecutive days within the period from October to December 2017. On the first day, the equipment for air samples was set up and started. Further, guidelines were given for hand wipes taken on the following day. On the following day, the air samples were collected together with the equipment, hand wipes as well as a surface wipe, taken on the wall. Further, the dust bag of the vacuum cleaner and the questionnaire were collected. All handling of samples were done with gloves.

According to the ethics approval by the Regional Ethics Committee (journal no H-16041946) and the Data protection Approval (SUND-2017-03) all participants were informed about their individual results, upon request. After individual consent, according to "General Data Protection Regulation", information of concentrations measured in the individual apartments was provided for the housing administration.

2.3. Air measurements

The air samples were taken in the living room. The sampling equipment was placed at least 1 m from the wall, avoiding direct sunshine, though taking into account the residents' ability to move around. The inlet of the sampling tube was placed approx. 1.3 m above the floor. The pumps were Gilair Plus, 2017 (Sensidyne, LP, USA) and placed in a sound-dampening box. The pumps were set to a constant flow rate of 2.0 l/min. At the beginning of a sampling period, the flow rate was manually checked with a ball flow meter (Porter, USA, Porter Instrument Company B–B17). Increasing temperature in the sound-dampening box within the first 3–5 h of the sampling period reduced the flow rate a few percent normalized to room temperature. The pumps were programmed to automatic stop after 24 h of sampling, though in some cases the air sampling was stopped manually shortly before the 24 h (sampling time range was 21–24 h). Air volumes collected were in the range 2.3–2.8 m^3 (volume logged by the pump, calculated to a reference temperature of 22 °C and a pressure of 101 kPa).The sampling tubes were "SKC tube 226–58" (SKC Inc., Eighty Four, PA, USA) with quartz filter and collection medium of a styrene-divinylbenzene resin (XAD-2) and polyurethane foam (PUF). The samples were collected as both gas phase and particle-bound PCB. The sampling tubes were fitted with a Teflon nozzle giving an inlet velocity of 1.25 m/s. After end sampling the Teflon inlet was removed, the sampling tube was plugged and placed in a Rilsan bag, which was sealed and stored in a freezer (−18 °C) until extraction and chemical analysis.

Table 1
Sampling periods for short-term measurements of air concentrations. Outdoor hourly temperatures taken from DMI.dk https://www.dmi.dk/vejrarkiv/.

home	Month/year	Sampling periods, start time	Sampling time, hours	Outdoor temperature range, hourly mean (min-max), °C
A	November 2017	10-14-18-22-02-02-10	4-4-4-4-4-4	−0.5 - 4
B	December 2017	16-22-02-06-16	6-4-4-10	7–8
C	November 2017	10-14-18-22-02-02-10	4-4-4-4-4-4	3–9
A	June 2018	10-14-18-22-02-02-10	4-4-4-4-4-4	10–16
B	June 2018	14-18-22-02-06-10-14	4-4-4-4-4-4	15–25

2.3.1. Short-term concentration measurements

In 13 homes, a 4-h air sample was taken in addition to the 24-h sample. The 4-h sample was taken during night-time, from 2 to 6 o'clock. The pump was programmed to automatically start and stop. Further, consecutive short-term measurements of the air concentrations were run in parallel with the 24-h sample in three exposed homes. In two of the homes the short-term measurements were repeated in June 2018. Starting time and sampling duration are given in Table 1, together with the range in outdoor temperature within the 24-h sampling period.

2.3.2. Chemical analysis

The chemical analysis of the air samples were carried out by Dansk Miljøanalyse A/S (Skelstedet 5, DK-2950 Vedbæk, Denmark). The whole sample, i.e. filter and adsorbing material, were extracted together in a mixture 1:1 (v/v) cyclohexane and acetone in an ultrasonic bath. The extracts were analysed by GC-MS (J&W 30 m DB-XLB, 0.25 mm ID, 0.25 µm film) with ^{13}C-PCB-202 as injection standard and dibromooctaflourobiphenyl as recovery standard. The seven indicator PCBs (PCB-28, -52, −101, −118, −138, −153, and −180) were analysed by a standardized accredited method. The expanded analytical uncertainty was 25%, but 35% close to the detection limit (LOD). LOD was 1 ng for the single congeners in a sample, i.e. a detection limit of typically 0.4 ng/m^3 for the daily samples. Further, the samples were analysed for PCB-8, -18, −31, −44, −66, −74, −99 and −105, in total 15 PCB congeners. These congeners were selected for better representation of the low-chlorinated congeners and were based on congeners in commercial mixtures as well as air of contaminated schools (Takasuga et al., 2006; Ampleman et al., 2015). Furthermore, the selection matched the analysis of blood and hand wipes that were also collected (Frederiksen et al., 2020). The total amount of PCBs in air samples was calculated as PCB_{sum7} multiplied by a factor 5. All blank samples were below the detection limit.

2.3.3. Temperature and relative humidity

Together with the air sampling, temperature and relative humidity were logged every 10 min with HOBO–U12-012 data logger (Onset, Bourne, MA, US). The logger was placed below the air sampling tube in a height of 1 m.

2.4. Dust

The dust bag from the participant's vacuum cleaner was collected at the second home visit. The dust bag was placed in a Rilsan bag and sealed. If the vacuum cleaner did not have a dust bag, dust was carefully transferred from the vacuum cleaner to a Rilsan bag and sealed. In two homes it was not possible to collect vacuum cleaner dust. The dust was stored in refrigerator until further handling. The dust was sieved with an automatic sieve shaker (Retsch 100, AS 200, Retsch GmbH, Germany). First, the dust bag was cut up along the edge and dust was taken from the dust bag over its entire length to obtain a mixture of old and younger dust. The dust was transferred to the top sieve. The dust was sieved in the following fractions: > 2 mm, 150 µm ≤ 2 mm, 75 ≤ 150 µm and < 75 µm. The fraction of < 75 µm was selected for analysis as this fraction is fairly homogeneous and represents the size range relevant for hand-to-mouth exposure due to adherence to skin etc. (Choate et al., 2006; Weiss et al., 2018). About 1 g of the fraction < 75 µm was extracted and analysed for PCB as described for the air samples. The detection limit was 10 ng/g for each congener in the sample.

2.5. Surface wipes

The wall in the living room was wiped following a pre-defined procedure. The wipe was a synthetic fibre tissue moistened with isopropanol (Alkoholswab 70% isopropanol, 30 × 60 mm, Mediq Denmark A/S, DK-2605 Brøndby, Denmark). The design of the wiper was inspired by one used by Clausen et al. (2016). In brief, a wipe was placed on a wooden piston covered with aluminium foil and held by a tube of stainless steel. The piston, with a diameter of 25 mm, was mounted on a rod with a spring. A wooden handle assured together with the spring a constant pressure of the wipe against the wall during use. The handle was protected from touching the wipe by the stainless steel tube. When wiping, the piston with wipe was pressed against the wall using the handle. Due to the spring, the piston pressed against the wall with a force of 10 N, equal to a pressure of about 20 kPa, when keeping the handle a few millimetres from the wall. A horizontal distance of 40 cm was marked on the wall by use of tape. The piston with wipe was moved above the tape across the wall at a constant speed of 40 cm in approximately 8 s. Thereby, a surface of 100 cm^2 was wiped. The procedure was repeated once for each wipe, in the same track and direction. It turned out that a majority of homes had sawdust wallpaper, which made the size of the wiped area more uncertain as the uneven surface caused the wipe to not fully touch the wall between sawdust bits. It was noted whether the wall was uneven or flat.

After wiping, the wipe was placed in a 10 mL vial (glass vial with screw top and steel cap with PTFE/butyl septa). The vial was placed in a Rilsan bag and sealed and stored in a freezer (−18 °C) until extraction and chemical analysis. Extraction and chemical analysis was done as described for the air samples. Blank samples, representing wipes mounted and dismounted on the piston in contaminated apartments, were all below detection limit.

2.6. Statistical analysis

Linear regression analysis and regression test were performed using Excel 2016. Box plots with interquartile range and whiskers of 5th-95th percentiles was performed using GraphPad Prism v. 8.0.2. Comparison of groups including levels of significance (Fig. 1) was done using Mann-Whitney *U* test in GraphPad Prism v. 8.0.2.

3. Results

3.1. Temperature

The mean indoor temperatures during the 24-h sampling period of air sampling varied between 18.3 °C and 26.3 °C. The variation in temperature during each measuring period was low as most of the sampling periods have a standard deviation of less than 1 °C (Fig. S2). The average of the mean temperatures were 22.0 °C.

3.2. Concentrations

Table 2 presents the mean, median and relative standard deviation (RSD) of the individual congener concentrations measured in air, dust and surface wipes from exposed and reference homes, respectively. PCB_{sum7}, PCB_{total} (= 5xPCB_{sum7}), and PCB_{sum15} are presented along

Table 2
The mean, median and RSD of the concentration of the individual congener measured in air, dust and surface wipe samples from exposed homes and reference homes, respectively, together with the detection frequency (%).

	Air								Dust								Surface wipes							
	Exposed (n = 53)				Reference (n = 20)				Exposed (n = 51)				Reference (n = 20)				Exposed (n = 53)				Reference (n = 20)			
	Mean ng/m^3	Median ng/m^3	RSD %	Detec-tion freq-uency %	Mean ng/m^3	Median ng/m^3	RSD %	Detec-tion freq-uency %	Mean ng/g	Median ng/g	RSD %	Detec-tion freq-uency %	Mean ng/g	Median ng/g	RSD %	Detec-tion freq-uency %	Mean ng/wipe	Median ng/wipe	RSD %	Detec-tion freq-uency %	Mean ng/wipe	Median ng/wipe	RSD %	Detec-tion freq-uency %
PCB-8	56	44	76	100	4	3	89	80	124	87	81	100	3	0.0	312	10	1	0.0	211	26				0
PCB-18	382	370	49	100	6	5	74	100	871	743	61	100	9	0.0	150	40	48	40	99	100	0.4	0.0	262	15
PCB-28	230	219	45	100	4	3	64	100	1598	1293	62	100	27	18	112	70	58	46	82	100	0.7	0.0	185	30
PCB-31	331	318	44	100	6	5	66	100	2063	1708	60	100	42	27	108	75	80	59	90	100	1	0.0	195	30
PCB-44	138	129	44	100	3	3	60	90	1563	1294	64	100	32	23	89	80	93	69	92	100	1	0.0	199	30
PCB-52	221	207	43	100	6	6	46	100	2645	2327	63	100	74	54	91	90	103	76	93	100	2	0.0	176	30
PCB-66	60	55	44	100	2	2	67	85	1174	978	71	100	23	11	147	55	71	53	95	100	1	0.0	203	25
PCB-74	29	26	44	100	1	0.8	87	75	674	556	68	100	12	0.0	180	40	29	20	104	98	0.4	0.0	245	15
PCB-99	8	7	55	100	0.3	0.0	156	35	210	187	63	100	9	6	116	50	9	6	115	92	0.1	0.0	261	10
PCB-101	12	11	49	100	1	1	108	55	464	397	66	100	62	45	103	85	18	13	97	98	0.8	0.0	214	20
PCB-105	3	3	64	92	0.0	0.0	447	5	79	64	83	96	4	0.0	164	30	5	4	112	75	0.5	0.0	260	15
PCB-118	2	2	72	89	0.1	0.0	313	10	200	169	71	100	20	15	104	70	8	4	127	89	0.4	0.0	254	15
PCB-138	0.3	0.0	144	40	0.1	0.0	308	10	66	61	58	98	47	42	76	85	2	1	133	60	0.5	0.0	218	20
PCB-153	1	0.9	87	70	0.2	0.0	308	10	96	85	60	100	76	74	81	85	3	3	119	66	1	0.0	222	20
PCB-180				0	0.0	0.0	447	5	25	23	72	84	29	20	108	70	0.3	0.0	325	11	0.2	0.0	345	10
PCB_{sum7}	466	433	43		12	10	54		5095	4258	61		335	273	77		193	149	90		5	0.0	177	
PCB_{total}	2330	2164	43		58	49	54		25473	21290	61		1674	1366	77		964	745	90		26	0.0	177	
PCB_{sum15}	1473	1463	44		33	31	56		11854	9988	62		469	386	80		529	416	91		10	0.0	178	

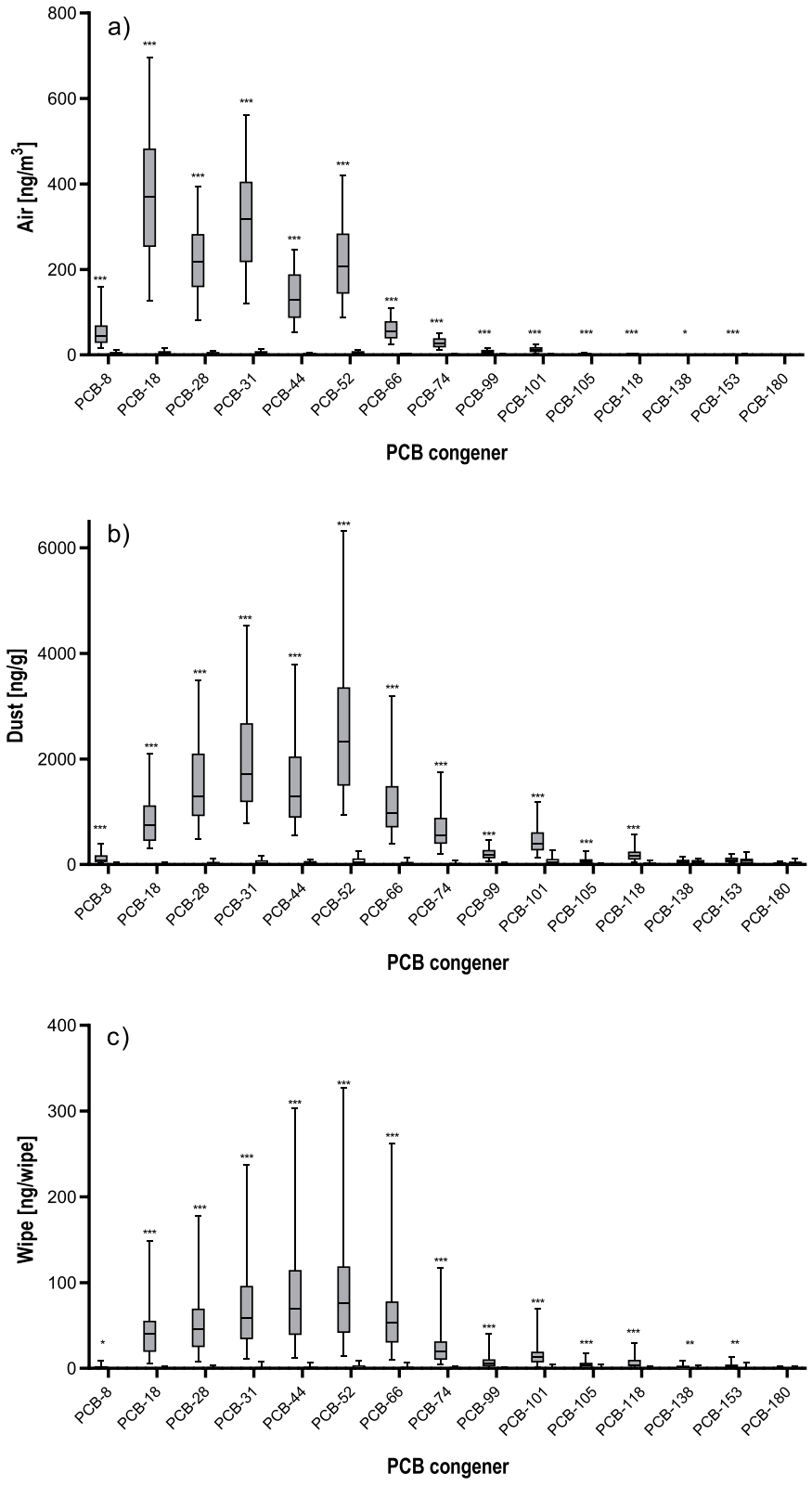

Fig. 1. Box plots with interquartile range and whiskers of 5th-95th percentiles of concentration levels in the exposed (grey) and reference homes (white) in a) air, b) dust and c) surface wipe (*p < 0.05, **p < 0.01, ***p < 0.001, Mann-Whitney *U* test).

with the detection frequency (%). Not detected (ND) congeners were given the value 0. Fig. 1 shows box plot with interquartile range and whiskers of 5th-95th percentiles of the single congeners in the exposed and reference groups for concentrations in air (a), dust (b) and surface wipes (c), respectively. Significance levels of differences between groups are indicated for the individual congeners.

3.2.1. Air

The air concentrations were elevated in the exposed homes, all lying above the Danish lower recommended action level of 300 ng PCB_{total}/m^3. This was expected and in 2019 there is an ongoing process handling the need for alleviation. All reference homes were below the lower recommended action level. In exposed homes, PCB-8 to PCB-101 were detected in all the samples. PCB-105 and PCB-118 were detected in more than 80% of these samples while PCB-180 was not detected at all in exposed homes. A similar pattern was observed in the reference homes, though with lower detection frequencies and only occasionally detection of PCBs with five or more chlorine. The congener concentrations were variating, with a RSD of more than 40% and highest within the reference group. The air concentrations had a significant difference in levels between exposed and reference homes. The levels were an order of magnitude lower in reference homes. The air samples were dominated by PCB-18 to PCB-52 in exposed homes.

3.2.2. Dust

In exposed homes all congeners but PCB-105, PCB-138 and PCB-180 (detection frequencies 96, 98 and 84%), were detected in 100% of the samples (Table 2). All congeners were detected in more than 80% of the samples. Of the 20 reference homes, the dust from two homes had no congeners found above the detection limit. In the remaining 18 homes, PCB-52 was observed in all samples while PCB-44, PCB-52, PCB-101, PCB-138 and PCB-153 were detected in more than 80% of the reference samples. The variance of the dust concentration was large (Table 2 and Fig. 1b), with a RSD for the single congeners at a minimum of 58% in exposed homes and for most congeners more than 100% in reference homes. In the exposed homes, the dominating congeners were PCB-18 to PCB-74. The levels of PCB-138 to PCB-180 in dust were similar in exposed and reference homes ($p > 0.05$), whereas a significant difference was observed for the other congeners.

3.2.3. Surface wipes

In exposed homes, PCB-18 to PCB-66 were detected in all surface wipes samples (Table 2) while PCB-8 and PCB-180 were the least frequently detected congeners. In the reference homes, 13 of the 20 samples were below the detection limit for all congeners. PCB-8 was not detected in any samples, while PCB-28 to PCB-66 were the most commonly detected. In exposed homes PCB-18 to PCB-66 were the dominating the concentration, with RSDs higher than observed for both air and dust samples (Table 2 and Fig. 1c). In the reference homes, all detected congeners were at a low level and to some extent dominated by PCB-28 to PCB-66. The RSDs were high and apart from the higher chlorinated PCBs in air also higher than the other media. Despite the large variations there was a significant difference between the single congeners in the samples from exposed and reference homes ($p < 0.05$), apart from PCB-180. Three parallel samples (same wall) from three different apartments showed a concentration RSD up to 60% for congeners constituting more than 5% of the sample, giving a rather low repeatability of the surface wipes, see SM, Table S1.

3.3. Correlations with air concentrations

The air concentrations (as PCB_{sum7}) were compared to the average indoor room temperature during the 24-h measuring period. Although high variations were seen, a significant positive correlation was observed in the exposed homes (Excel regression, $\ln(PCB_{sum7})$ versus temperature, $R^2 = 0.2$, $p < 0.05$). Fig. 2 presents the air concentrations (log10 scale) versus temperature. Comparing concentrations of PCB_{sum7} in dust with air concentrations in exposed homes, a significant positive correlation was observed (Excel regression, $R^2 = 0.3$, $p < 0.05$), though the variability was high (Fig. 3). No significant correlation (Excel regression, $R^2 = 0.04$, $p > 0.05$) was observed comparing the results of the surface wipes and air concentrations in exposed homes. Fig. S6 in SM shows concentrations of PCB_{sum7} in surface wipes versus air, divided in exposed and reference homes.

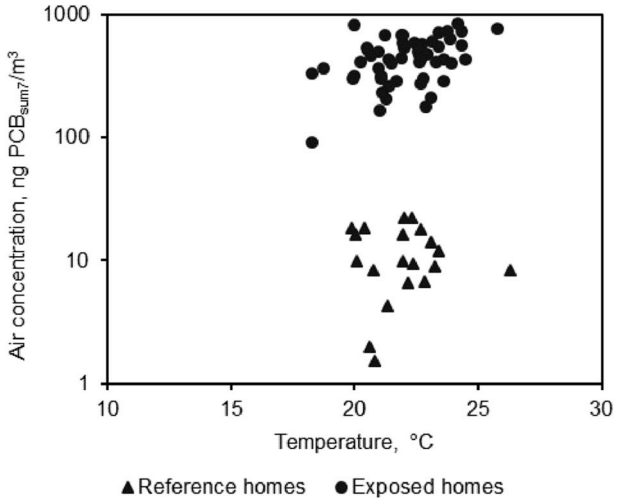

Fig. 2. Concentrations of PCB_{sum7} in air versus the average room temperature in exposed (circles) and reference (triangles) homes (one outlier excluded).

3.4. Diurnal variability in air concentrations

3.4.1. Short-term concentration measurements

Consecutive short-term measurements of the air concentrations covering all 24 h were run in parallel with the 24-h sample in three exposed homes. Fig. 4 shows the measured short-term concentrations and the 24-h mean value of PCB_{sum7}. Figure a, c and e present results from three apartment visited during the investigation in November and December 2017. Figure b and d show results from two of the apartments revisited in June 2018. The average room temperatures in the short-term periods are also shown in Fig. 4. In general, small variations were seen for both short-term concentrations and temperature over time. Within the 24-h period the short-term air measurements in the five data sets varied between 5 and 25% RSD.

Concentrations and indoor room temperatures measured in home A and B in November/December 2017 and June 2018 were at the same level. The deviation between the 24-h values were less than 15% (difference compared to average). Most of the measuring periods had rather low variations in hourly mean values of the outdoor temperature (Table 1). The largest temperature difference outdoor (10 °C) was

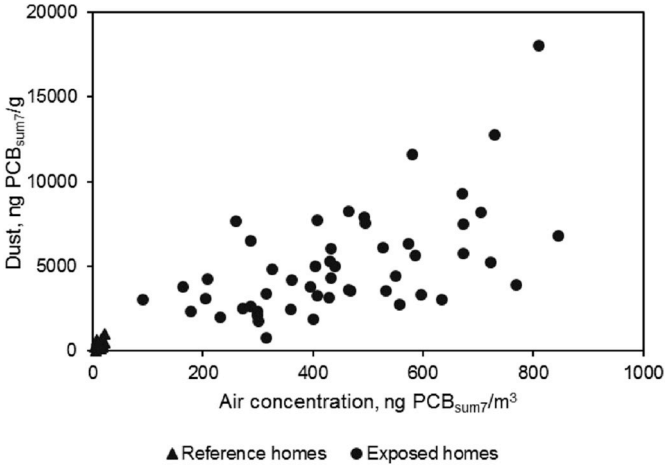

Fig. 3. Concentrations of PCB_{sum7} in dust versus air in exposed (circles) and reference (triangles) homes.

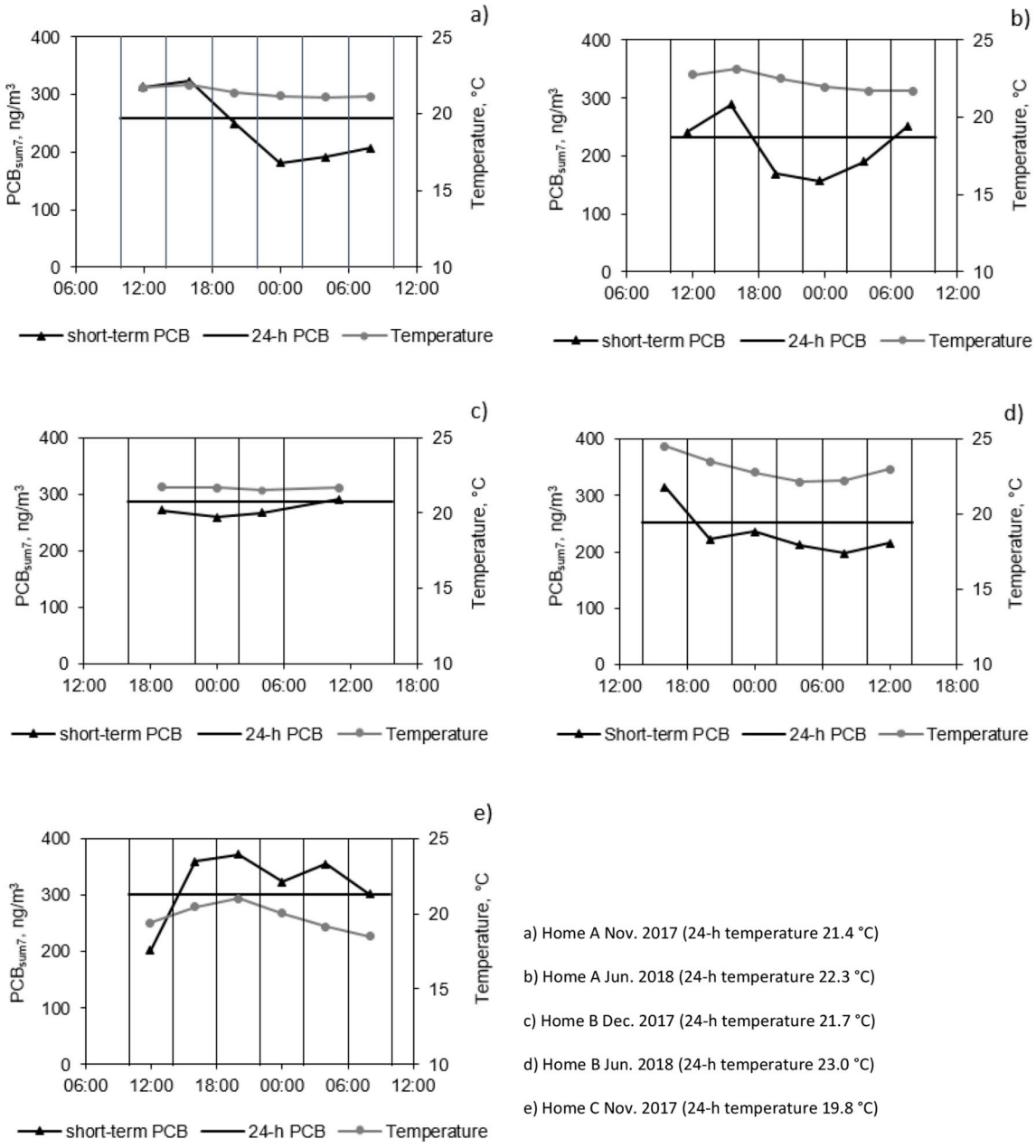

Fig. 4. Air concentration of PCB_{sum7} (short-term and 24-hr) together with temperature (averaged to short-term period) in three homes winter 2017 and repeated in two homes June 2018.

observed during the measurements in home B in June 2018, though it did not result in large variations in temperatures indoor.

3.4.2. 4-h measurement during the night

Fig. 5 shows the air concentrations (as PCB_{sum7}) in 13 homes measured as 4-h values during the night (2–6 am) versus the 24-h values together with the linear regression line. No significant difference was observed determining air concentrations as 4-h night-time sampling compared to the 24-h sampling. The variation in temperature within the 24-h periods was low (RSD < 5%).

3.5. Pattern of congeners

The pattern of congeners were calculated as the concentration of the single congener relative to the sum of congeners, i.e. the relative distribution (%). Only congeners detected in minimum 80% of the samples were included within the three respective media in exposed as well as reference homes. No congeners in surface wipes from reference homes meet this criterion. In two dust samples from reference homes, no congeners were detected and they were left out of the calculation. Further, a sample with only PCB-52 detected was left out. Fig. 6a, b and c show box plot of the relative distribution measured in exposed and reference homes for air, dust and surface wipes, respectively. Although the repeatability of the concentrations of the surface wipes was rather poor, the sampling method showed a good performance reflecting the pattern of the congeners in samples of wall paint, see SM.

In the exposed homes, small variations in the pattern of congeners were observed for the respective media having a rather uniform pattern (Fig. 6). Congeners amounting to more than 5% of the sum had a RSD of less than 30% for the single congeners. Compared to the exposed homes, the reference homes showed far more variation (Fig. 6). In exposed homes the general pattern of congeners in all three media were dominated by tri- and tetraPCBs, though with an air pattern differing from that of dust and surface wipes, being more alike. The air samples

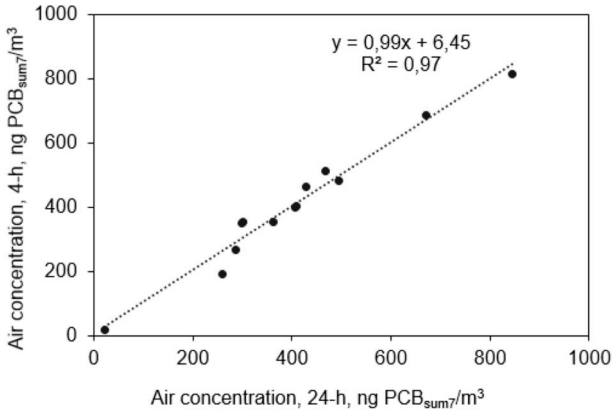

Fig. 5. The air concentration (as PCB_{sum7}) in 13 homes based on 4-h sampling at night (2–6 am) versus results based on 24-h sampling. The linear regression line is shown.

were dominated by PCB-18 and PCB-31, while the dust and surface wipes were dominated by PCB-28 to PCB-52. Of the higher chlorinated congeners, PCB-101 concentrations were more prevalent in dust and surface wipes compared to air.

4. Discussion

4.1. Concentration levels

4.1.1. Air

Looking at the average value of PCB_{total}, the level of exposed homes was 40 times higher than the reference homes. The average value (2330 PCB_{total}/m^3) of exposed homes in this study are higher than the average level (1030 ng PCB_{total}/m^3) observed by Frederiksen et al. (2012) in another Danish housing estate with indoor PCB containing sealants. The levels are higher than observations from Sweden, where Sundahl et al. (1999) found up to 600 PCB_{total}/m^3 and Corner et al. (2002) found up to 270 ng PCB_{total}/m^3, though both in an apartment block with outdoor sealants as the primary PCB source. Reports from German schools and public buildings show large variation and maximum levels in single rooms exceeding the levels we observed (Benthe et al., 1992; Liebl et al., 2004; Gabrio et al., 2000). Kohler et al. (2002) report levels up to 4200 ng PCB_{total}/m^3 from public buildings in Switzerland, levels which are comparable to our findings. MacIntosh et al. (2012) report levels up to 1800 ng/m^3 in indoor air in US schools, though these concentrations are based on analysis of homolog groups and therefore not directly comparable with concentrations based on PCB_{sum7}. Kraft et al. (2018) analysed mono-, di- and trichlorinated PCBs in the indoor air of 35 contaminated offices. The total sum of the measured PCBs showed a median of 479 ng/m^3, with PCB-31, PCB-18 and PCB-28 dominating the trichlorinated PCBs (median levels of 39, 31, and 26 ng/m^3, respectively). PCB-8 was the most prevalent dichlorinated congener with a median value of 9.1 ng/m^3. These values are considerable lower than the median values measured in this study.

The reference homes had an average concentration of 58 ng PCB_{total}/m^3, though with a large variation. These results are comparable to other Danish findings in homes done by Frederiksen et al. (2012) and Gunnarsen et al. (2009).The levels observed were higher than an average of 3 ng PCB_{total}/m^3 in 31 homes in UK reported Harrad et al. (2006). Hazrati and Harrad (2006) observed, that buildings erected in the period with use of PCB in building product (1950–1979) had significantly higher PCB concentrations indoor than buildings erected later.

4.1.2. Dust

For exposed homes, the mean dust concentration of PCB_{sum7} (5095 ng PCB_{sum7}/g) is in the upper interval reported by Corner et al. (2002), who found variating levels of PCB_{sum7} in the range (0.01–5.4 μg/g) in dust from apartments with PCB in outdoor sealants in Sweden.

For the reference homes, the mean dust concentration (335 ng PCB_{sum7}/g) were slightly higher than the levels observed by Gunnarsen et al. (2009), reporting levels up to 124 ng PCB_{sum7}/g for a limited number of homes, having air concentrations of less than 30 ng PCB_{total}/m^3. Harrad et al. (2009) report concentrations of PCB in domestic dust from USA and Canada with average values of 220 ng/g and 290 ng/g, while in the UK and New Zealand, they observed lower values, 110 ng/g and 67 ng/g, respectively. Their samples were taken with a vacuum cleaner following a specific procedure and the level of PCB was determined as the sum of PCB_{sum7} and PCB-31 and -105. Our mean value for this sum of congeners were 381 ng PCB/g and comparable to the results from USA and Canada. Knobeloch et al. (2012) collected dust from residential vacuum cleaner bags from 20 homes in Wisconsin, USA. The samples were sieved to < 1 mm and analysed for 60 congeners. They report levels between 8 and 1187 ng/g, with PCB-101, -110, −138 and −153 found in all samples and together with PCB-95 comprising more than a third of the total. This is similar to our results having a detection frequency of 85% for PCB-101, -138 and −153, in average comprising almost 40% of PCB_{sum15}. Knobeloch et al. (2012) found the highest levels in homes built from 1959 to 1970, the same time period as all our reference homes represent.

Looking at the highly chlorinated congeners (PCB-138, PCB-153 and PCB-180), the results from exposed and reference homes showed similar levels. In a survey in four areas of the USA with collection of a large number of vacuum cleaner bags with following sieving to 150 μm, Colt et al. (2005) analysed for the highly chlorinated congeners PCB-105, PCB-138, PCB-153, PCB-170 and PCB-180. Of about a thousand dust bags, a little less than half had detectable amount of the congeners. Of the detected samples 2/3 had a concentration below 200 ng/g. Though we did not analyze PCB-170, we observed similar levels (Table 2).

4.1.3. Surface wipes

Surface wipes are used as performance evaluation of mitigation (U.S. Environmental Protection Agency, 1991) and also found suitable for dust sampling on hard surfaces (Cettier et al., 2015), though we have not found any investigations using surface wipes for detection of PCB on vertical surfaces similar to the current approach on walls. The amount of PCBs on the surface wipes were highly variating, but a significant difference was observed between exposed and reference homes.

4.2. Pattern of congeners

The air will be enriched in the less chlorinated congeners compared to the primary source (Balfanz et al., 1993; Kohler et al., 2002; Heinzow et al., 2007; Guo et al., 2011). Therefore, factors relating to the type of PCB mixture and degree of chlorination as well as the concentration in sealants, position and surface area will influence the degree of indoor air contamination. The air samples from the exposed homes showed a rather uniform pattern of congeners with tri- and tetraPCBs dominating the content. This could reflect the use of a specific product of PCB as source for the contamination. Samples of sealants from two apartments in different high-rise buildings showed a congener pattern comparable to the commercial PCB products Aroclor 1248 and Clophen A-40 (see SM, Fig. S7) indicating that one of these products likely have been the additive in the sealants.

The indoor air of the reference homes showed detectable amounts of PCB and to some extent with a similar congener pattern as the exposed homes. The dominating sources to PCB in reference homes can have origin in legacy produced industrial products used in a range of applications. In Denmark, Grontmij & COWI (2013) made a survey of PCB in building materials from buildings erected in the period 1950–77.

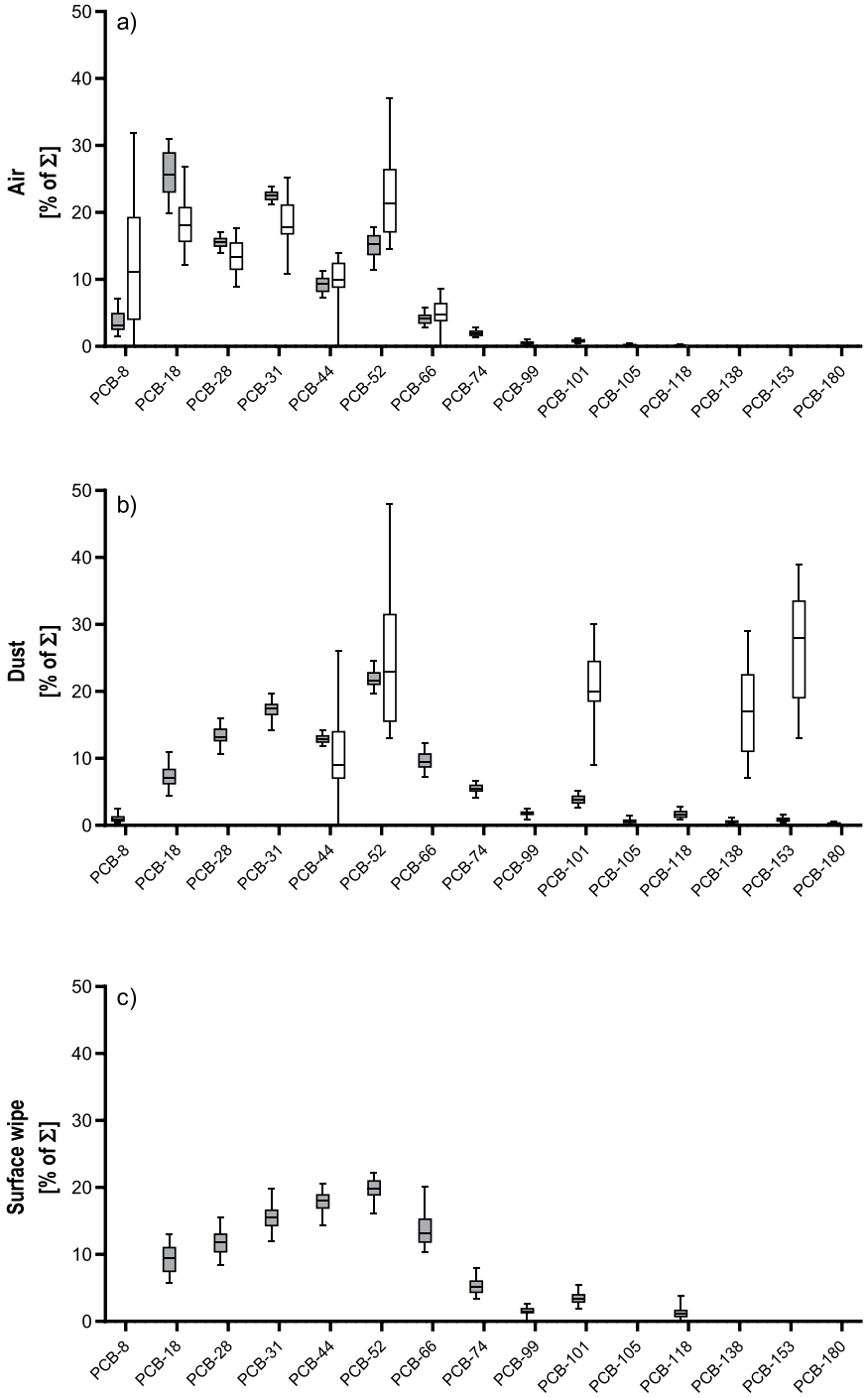

Fig. 6. Box plots with interquartile range and whiskers of 5th-95th percentiles of single congener relative to the sum (%) for congeners in the exposed (grey) and reference homes (white) in a) air, b) dust and c) surface wipe with minimum 80% detection frequency and more than one congener detected.

Materials with low levels of PCB were found a majority of the buildings, often due to low content of PCBs in paints. Some PCB congeners found in pigments do not appear to be derived from traditional commercial products such as Aroclor. PCB-11 can be formed inadvertently by the production of certain yellow pigments (Rodenburg et al., 2010; Shang et al., 2014), and studies have shown different PCBs in certain azo and phthalocyanine pigments used in paint (Hu and Hornbuckle, 2010). Vorkamp (2016) have reviewed inadvertent formation of PCB-11 and other PCB congeners and their occurrence in consumer products and in the environment. The detection of PCB-11 in the polar environment indicates long range atmospheric transport. It was our intention to include PCB-11, but it co-elutes with PCB-18 on the chosen column. Unfortunately, PCB-18 give rise to a low signal in the MS-track of PCB-11, thus the presence of high levels of PCB-18 in the samples cause large uncertainties in the determination of trace levels of PCB-11.

A uniform pattern were also observed in samples of dust and surface wipes from exposed homes. It is probably reflecting the uniform pattern of congeners in air as we expect the air to be a major contributor to the contamination of both dust and surface on the wall. There is a clear difference in congener pattern of air samples compared to dust and

surface wipes as expected due to the physical chemical properties of the single congeners. As shown by Weschler and Nazaroff (2010) the octanol-air partition coefficient of an SVOC is often a strong predictor of its abundance in settled dust relative to its gas phase concentration. The air samples are dominated by PCB-18 and PCB-31, while the dust and surface wipes are dominated by PCB-28 to PCB-52. Relative, PCB-44 seem to behave differently in dust samples compared to surface wipes. Of the more chlorinated congeners both dust and surface wipes have relatively more PCB-101 compared to air. Looking at the congener pattern of dust from reference homes, it is different from the exposed homes. The variation was larger and higher chlorinated PCBs were more dominating.

4.3. Influencing factors

4.3.1. Air

In this study, we observed variation in the air concentrations of PCB in the exposed homes (Table 2, Fig. 1). Our data show some correlation with the room temperature, though only with a small degree of explanation. Lyng et al. (2016) showed a strong impact of temperature on gas concentration at near steady-state conditions; raise in temperature from 20 °C to 24 °C caused an increase in the partial pressure of 50–60% for the congeners PCB-28, PCB-52 and PCB-101. There are numerous other factors influencing both concentration levels and composition of congeners of PCB in indoor air (as discussed above). Variations in sealant concentrations, placements and maybe degree of spill can cause some of the variation observed in this study. Balfanz et al. (1993) point out, that PCB often was added to the sealant materials immediately prior to their use, leading to great diversity in content and sometimes as mixtures of different commercial products too. Frederiksen et al. (2012) observed the use of different commercial products through an estate. Further, outdoor as well as indoor temperature, humidity and air exchange rate are observed to have an impact (Balfanz et al., 1993; Kohler et al., 2005; Lyng et al., 2015 & 2016). We saw no difference in air concentration levels between a winter and a subsequent summer visit in two homes, though again the indoor temperature was rather stable and similar in the two seasons. Apparently, outdoor temperature did not have a large influence on the indoor air concentration, which could be expected, as the sources were indoors and maybe influenced by the central heating system; however the number of samples covering the seasonal variation is limited. We do not know the influence of outdoor and indoor temperature differences on the air exchange rates, though would expect a higher air exchange rate in summertime.

We observed limited diurnal variations in air concentrations within a subgroup of homes with extended sampling (Figs. 4 and 5). It could be a result of a rather stable indoor room temperature during the sampling periods together with the dynamic behaviour of the tertiary sources. During the period of sampling the apartments were in normal use and increases in air exchange rate and with that a dilution of PCB from the strong sources may be counteracted by desorption of PCB from interior surfaces as observed by Lyng et al. (2015).To our knowledge, no information about diurnal variability of the PCB air concentrations in exposed homes during everyday use has previously been reported.

Prior to this investigation, the housing administration had initiated air sample measurements conducted by professionals within the period 2010 to 2016 (Golder Associates A/S, 2017). In comparison, our measurements were at the same level or above in apartments with prior results. The differences observed could be due to several of the factors already mentioned to influence the air concentrations, though also the fact, that different procedures and chemical laboratories have been used. Another issue is whether the high-rise buildings, constructed more than 40 years ago, have reach a certain steady state between sources, air concentrations and interior surfaces or whether the concentration levels in air and interior surfaces still are increasing. This is also a subject of consideration estimating long term exposure levels.

4.3.2. Dust samples

The exposed homes in this study showed a significant positive correlation between air and dust concentrations, though with a great scatter (Fig. 3). It opens up for a possibility to use vacuum cleaner dust as a screening tool for elevated air concentrations of the less chlorinated PCBs in the indoor air. It is not suitable for evaluating the air levels compared to the Danish recommended action levels, though could be a method simplifying the identification of houses that ought to be examined closer. The dust bag could be shipped to the laboratory by the residents themselves and would save the cost of having a visit of a professional taking an air sample before any indications are identified. However, more knowledge is needed concerning dust in buildings contaminated with lower levels of PCB and also whether a dust analysis would reveal contamination from high-chlorinated technical products of PCB. In an investigation with sampling in nine buildings in Denmark, Wilkins et al. (2002) report, that shelf dust collected in one building revealed a large amount of tri- to pentaPCBs, 20 times the amounts found in the other buildings. This was attributed to sealants containing PCB, indicating that elevated concentrations of the tri- to pentaPCBs in dust could indicate a possible PCB source, which might elevate indoor air concentrations.

4.3.3. Surface wipes

The concentration levels of the surface wipes are varying between exposed homes, though there was a rather uniform pattern of the congeners (Fig. 6c) with domination of tri- and tetraPCBs. This must likely reflects the indoor air as source for the wall contamination, though not with a quantitative relation. As pointed out by Weschler (2016) occupants transfer skin oil to and shed skin flakes onto indoor surfaces. We expected a wipe on the wall to collect a complex mixture of these organic compounds originating from the residents, together with PCB and other deposit of different origin (e.g. cooking, smoking) as well as residues from wall paint. Although no correlation between PCB_{sum7} in air and wipe was seen, a significant difference was observed between exposed and reference homes and the results indicate that the surface wipes have a potential as a screening tool as also discussed for the dust. Further development should include homogeneity of surface roughness to eliminate differences in wiped area and an increase the wiped area to increase detection frequency.

4.4. Strengths and limitations

A strength of the study was the analysis of the many congeners relevant for indoor air contamination with PCB. The range of analysed congeners showing uniform patterns in both air, dust and surface wipes in exposed homes points to a single product as original PCB source, though at the same time gives resilience to the data set collected. As part of an investigation of the total indoor exposure of residents in contaminated buildings and with the range of congeners analysed in samples of blood and hand wipes too, the results gives a good basis for further data analysis of the relative importance of different exposure pathways.

Another strength of the study was the similar building characteristics of the exposed homes, representing four types of apartments, located on identical floorplans in five high-rise buildings. Further, the participants of the study have answered a questionnaire giving information of among other things their airing and cleaning behaviour. This information, together with the similar building characteristics and the resilient data set obtained, gives us a good starting point for further data analysis of factors with impact on the air concentration and with that more knowledge of relevance for alleviation and remediation.

A limited number of observations weakens some of the results in the study. It was a small reference group and it should be regarded as a reference to the exposed homes rather than representing any general background level. Seasonal variations in air concentrations in exposed homes are based on a comparison of results revisiting only two homes

the subsequent summer. It is a spare basis for conclusions, though the results do not conflict with our hypothesis of tertiary contaminations of interior surfaces being able to counteract changes in indoor air concentration, despite possible decrease in source strength of the sealant source and an expected increase in air exchange rates during summer time.

Another limitation of the study is the results of the surface wipes being highly variable and to some extent this might be due to the method itself. It is speculated whether the large variation between tests with parallel samples is caused by a rough surface making the wiped surface area variable. As a majority of the homes had sawdust wall paper, which has a rough surface, the wiped surface area might be less well defined. Further, factors like time since last painting, type of paint, smoker/non-smoker home etc. might influence the wipes and the absolute levels should be interpreted with caution.

5. Conclusion

This study confirm the presence of buildings with highly elevated indoor air concentrations of PCB and therefore a need for identifying and alleviate PCB in buildings causing a health concern to the occupants. The 53 apartments located in high-rise apartment buildings with indoor sealants containing PCB showed significant elevated total concentrations of PCB in air, dust and surface wipes compared to 20 reference homes. On average, the air concentration of PCB_{total} was 40 times higher in exposed homes compared to reference homes, while dust and surface wipes showed 25 and 50 times higher levels, respectively. Variations in air concentrations in exposed homes was to a small extent explained by differences in indoor room temperature. Additional short-term air measurements in a subgroup of exposed homes revealed limited diurnal variation in air concentrations. The results were obtained during everyday use and the small variations could be due to small variations in room temperature as well as interior contaminated surfaces counteracting changes in air concentrations by desorption of PCB.

In exposed homes, all three media were dominated by the tri- and tetraPCBs and showed a rather uniform congener pattern, though with a different pattern for the air samples compared to dust and surface wipes, which were more alike. The uniform congener pattern in the air most probably reflect a single PCB product as dominating source in the sealants emitting PCB to the air. From the air PCB are sorbed to surfaces and dust, redistributing PCB from the original source to all interior surfaces.

In the reference homes the air concentration were dominated by the less chlorinated PCBs, while for dust it was less distinct. Only a third of the surface wipes showed detectable amount of any congener.

The levels in air and dust samples were significantly correlated for PCB_{sum7} in exposed homes, however with a great scatter. There was no significant correlation observed between air concentrations and surface wipes, though there was a significant difference between surface wipes from exposed homes compared to the reference homes. The results show that both dust and surface wipes have the potential as a screening tool for identifying elevated air concentrations of the less chlorinated PCBs.

Declaration of competing interest

The authors declare that they have no conflicts of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ijheh.2019.113429.

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