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Indoor air concentrations of PCB in a contaminated building estate and factors of importance for the variance

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ABSTRACT

The paper presents results of statistical analyses of indoor PCB air concentrations related to building characteristics, time trends and resident behavior. More than 150 measurements of air concentrations were made in five high-rise apartment buildings with PCB containing sealants in an estate in Denmark. The indoor air concentrations consists of two datasets. Data set A are short-term nighttime measurements in both kitchen and living room taken during the period from 2010 to 2017. Data set B consists of 24-h measurement taken in the living room late 2017. Data set B also holds information from a questionnaire of among other things cleaning habits filled out by the residents and concentrations of PCB in dust from the vacuum cleaner bag of the homes. Both data sets show higher air concentrations associated with higher indoor temperature. Data set A further show, that this was associated with wintertime and it is speculated, that the higher air concentrations in cold weather are due to underfloor PCB sources being heated by radiator pipes. Building, floor level and apartment type were used as variable as the apartments were very similar and the type included many characteristic parameters. No relations were seen between air concentrations and these parameters. Repeated measurements of air concentrations from a number of apartments between 2010 and 2017 showed some variation over time, though no clear tendency was observed. Frequent cleaning habits had a minor influence on lowering the air and dust concentrations.

1. Introduction

Polychlorinated biphenyls (PCBs) are aromatic, synthetic chemicals and many of the PCB congeners are highly persistent and accumulate within food chains [1]. The technical properties of PCB has resulted in numerous commercial applications, including dielectric isolators in electrical equipment and addition to polymeric building materials as plasticizer or flame retardant [2]. PCB is a global environmental problem and among the initial 12 Persistent Organic Pollutants in The Stockholm Convention, recognized as causing adverse effects on humans and ecosystems. The International Agency for Research on Cancer has classified PCB as carcinogenic to humans (Group 1) [3]. Most countries have joined the Stockholm Convention, which came into force in 2004. It 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 [4]. Some uses of PCB have shown a potential to contaminate the indoor air in buildings significantly (e.g. Refs. [5–9]. The contamination of the air in buildings with PCB containing sealant relates to a range of factors, often specific for the individual building and even on room level. The indoor air will be enriched in the less chlorinated congeners compared to the primary sources [5,10,11]. The type of PCB mixture, degree of chlorination and concentration in sealants as well as their position and surface area will influence the indoor air contamination as will factors like temperature and air exchange rate [5,6]. In a Danish survey, Grontmij & COWI [8] observed elevated air concentrations in buildings having indoor sealants containing more than 10%

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(w/w) PCB. Other sources like PCB containing paint and PCB leaking from capacitors in fluorescent lighting ballasts also caused elevated concentrations, though no clear relations were found between material and air concentrations [8]. An intervention study demonstrated a strong relationship between indoor temperature and air concentrations and this was supported by field data [12]. Other investigations have found more or less pronounced relations to indoor temperature [5,6,9,13]. Outdoor temperature or its influence on parameters like air exchange rates and wall temperatures might have an impact on the air concentrations [5,14]. The influence of changes in air exchange rate can differ significantly, depending on the position of sealants indoor or outdoor and pressure conditions in the building [15,16].

PCB belongs to the group of Semi-volatile Organic Compounds (SVOC). The physical-chemical properties enable these compounds to evaporate from the primary sources to the indoor air and from the air sorb to all interior surfaces, causing a widespread contamination and redistribution of sources [17,18]. The contaminated surfaces becomes a “tertiary source”. PCB in sealants can also migrate to adjacent building materials, making these “secondary sources”. The tertiary sources have the ability to sorb as well as desorb SVOCs, depending on the conditions. These dynamic properties can cause great challenges during alleviation and remediation of PCB contaminated buildings as these tertiary sources can have the strength to maintain an unsatisfactory level of PCB in the air for a very long time, despite removal of primary and secondary sources. A study from a Danish building estate, conducted after a preliminary alleviation including cover of indoor PCB containing sealants, showed indoor air concentrations influenced by the surface (area of ceiling and walls) to volume ratio in apartments, indicating an influence of the tertiary sources [13]. Further, the air concentrations related to the indoor temperature and for the less volatile congeners (≥5 Cl) also related to the self-reported frequency of vacuuming [13].

The present study explore data collected in a PCB contaminated part of the building estate “Brøndby Strand Parkerne” (BSP) in Denmark. The estate was built in the period 1969–74 and the use of PCB was ceased throughout the process of constructing the estate, i.e. the buildings erected earliest contain PCB in the sealants whereas the later ones do not (see below). In the contaminated part, the estate owners have initiated investigations measuring air concentrations in more than 100 apartments in the period 2011–2017 and the contamination was assessed to concern almost 300 apartments. Late 2017 an investigation in the estate collected samples in 53 contaminated apartments [9]. Elevated air concentrations of PCB were dominated by the lower chlorinated congeners and the exposure have been documented to raise serum levels of these congeners within the residents in BSP [19] as well as in other buildings [20,21]. The exposure to contaminated indoor environment and the toxicity of the lower chlorinated congeners are less well studied. From an epidemiological perspective, a comparison of exposed and non-exposed residents could be a future possibility to gain more knowledge of the potential health effects related to indoor PCB exposure.

The purpose of the study was to analyze relations between air concentrations of PCB and parameters related to building characteristics and resident behavior in order to improve the ability to advise on how to reduce exposure of residents and the basis for remediation planning. Furthermore, the aim was to provide a scheme for gap-filling of exposure data in an epidemiological study of effect of living in PCB-contaminated housing.

2. Material and methods

This study analyzed two datasets: Data set A and B. Data set A consists of data collected on the initiative of the estate owner to investigate the extent of the PCB contamination of the indoor air in the buildings, in this context named “Investigation A”. Data set B was collected as part of a research project investigating the total indoor exposure of residents in PCB contaminated apartments in the building estate (“Investigation B″). Results of PCBs in serum and hand wipes from exposed residents and a reference group are presented in Frederiksen et al. [19]. Details of building description and obtained air concentrations of data set B are given Andersen et al. [9].

2.1. The buildings

Below is a building description based on Andersen et al. [9] together with information relevant for the statistical analysis. The building estate consists of 12 almost identical 15-story apartment buildings in Brøndby Strand Parkerne in Denmark, erected in 1969–1974. The buildings are placed in groups of three, lying on an east-west line, with the groups being approximately 400–800 m apart. The buildings are facing the compass directions the same way and with the same initial floor plan, i.e. the individual types of apartments have the same orientation and are exposed to sun and wind from the same direction in all high-rises. Floor plan is shown Fig. S1. The five buildings erected first, have PCB in sealants around the light façade elements indoors and outside on the glazed balcony and some windows [22,23]. 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 containing sealant on the concrete slabs below the wooden floors was observed as an additional source.

Air samples in investigation B were analyzed for 15 congeners. The samples showed a rather uniform pattern of congeners, having tri- and tetra chlorinated PCBs dominating the content. This could reflect the use of one specific product of PCB as source of the contamination. A few samples of sealants, collected in two apartments situated in different buildings, showed comparable composition of congeners and in average a content of 15% (w/w) PCB_{total} (RSD 12%, n = 7). The congener pattern was comparable to the commercial PCB products Aroclor 1248 and Clophen A-40 (see SI, [9]).

The construction principle of the high-rises 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 floorplans are identical, comprising four apartments in sizes 64, 77, 103 and 129 m^2. These areas are the official Danish Building and Housing Register (BBR) areas measured to the outside of walls and including fractions of shared floor areas connecting lifts and stairs with main doors to apartments. A few of the 129 m^2 apartments have been divided into two apartments. However, there were only few of those apartments investigated (five in Data set A and two in Data set B) and they were excluded from the final regression analysis due to small sample size. In data set B further two apartments did not match the used categories and was excluded from the statistical analysis of building characterization. The 129 m^2 apartment differs from the other apartment types by having a kitchen facing a concrete façade without a balcony. There are elastic sealants outdoor around the windows in the concrete façade and few samples have shown low content of PCB [24]. The kitchens in the other apartments face the light façade elements with indoor PCB containing sealants along floors and wall (Fig. S1). The apartment type was characterized by the floor area and ratio of surface area to volume as well as length of sealant to volume. The calculated parameters were based on measures made on building drawings. The volume of the apartment, living room and kitchen were calculated from the floor area times the height to the ceiling (2.5 m). The surface area was calculated as the summed area of floor, ceiling and walls, including doors and windows. Built-in closets and width of walls were disregarded. We have not attempted to estimate the surface area of furniture, carpets etc. as we found this too complicated and uncertain as every home is different. The floor levels of the apartments were divided in three groups: 1–5, 6–10, 11–15.

There is central mechanical exhaust ventilation connected to kitchens and bathrooms. The buildings have central heating supplied from one central installation shaft in each apartment. Connecting pipes are led under the wooden floors to the radiators (under most windows), where also some of the sealants are situated (see SI, [9]). In our analysis the heating season is defined as "winter" covering the month November to March, whereas "summer" is covering the month April to October, expecting no central heating demand.

2.2. Data set A

2.2.1. Air samples

Indoor air concentrations of PCB were measured over a period of 6 years (June 20, 2011–June 26, 2017) in BSP. In total 114 apartments were visited. Air samples were obtained by active filtration over filter and sorbing medium with a flow of 2 l/min by use of time programmed SKC AirCheck Sampler - Model 224-PCXR8. Sampling media composition and volume as well as time of collection were changed over the years due to change in official guidelines (Tables S1 and S2).

At least 8 h prior to sampling apartments were vigorously ventilated for 10–15 min by a measurement technician. Residents were told to keep windows and external doors closed until the next morning and otherwise act as usual. Two parallel samples were collected in each apartment during nighttime, one sample in the kitchen and one in the living room, all together 222 samples were made. The samplers were delivered by the commercial laboratory Eurofins A/S, who also analyzed the samples. The whole sample, i.e. filter and adsorbing material, were extracted together. The seven indicator PCBs (PCB-28, -52, −101, −118, −138, −153, and −180) were analyzed by GC-MS with a standardized accredited method. The expanded analytical uncertainty was about 25–35%, though higher for PCB-180. LOD was 0.2 ng/sample for the single congeners. Results are presented as the sum of the seven indicator PCBs, PCB_{sum7}.

14 apartments were revisited with repeating measurements in living room and kitchen in investigation A.

2.3. Data set B

2.3.1. Air samples

The air concentrations were 24-h samples taken in the living room within the period October to December 2017. The residents were asked to behave as usual during the period of air sampling. There was no prior airing out and no restrictions to airing during the sampling. The samples were analyzed for the seven indicator PCBs and eight other PCB congeners. Details are described in Andersen et al. [9]. The results are presented as the sum of the seven indicator congeners, PCB_{sum7}, as well as the sum of analyzed congeners in homolog groups (2 + 3 Cl: PCB-8 to −31; 4 Cl: PCB-44 to −74; 5 Cl: PCB-99 to −118; 6 + 7 Cl: PCB-138 to −180).

2.3.2. Dust samples

Dust samples were the sieved fraction (<75 μm) from collected vacuum cleaner bags from 51 exposed homes. The samples were analyzed for the seven indicator PCBs and eight other congeners (the same as the air samples). Details are described in Andersen et al. [9]. The results are presented as the sum of the 15 analyzed congeners, PCB_{sum15}, as well as PCB_{sum7} and the homolog groups mentioned above.

2.3.3. Questionnaire

In investigation B the homes were visited on two consecutive days when taking the air samples. The residents received a questionnaire the first day and was asked to fill out and return it the next day. The questionnaire included among other things detailed information on cleaning frequencies defined as vacuuming, dusting and floor washing as well as airing routines. "Airing" was defined as venting by making draft in the wintertime.

2.4. Effect of time

In investigation A, 14 apartments were revisited with repeating measurements in living room and kitchen. In addition, 17 apartments within investigation A were also visited within investigation B. As described in Andersen et al. [9] eight of these apartments had additional 4-h measurements made during night-time, showing no significant difference to the 24-h measurements. Further, limited diurnal variation was observed within a subgroup of homes with extended short-term sampling. It could be a result of a rather stable indoor room temperature during the sampling periods together with the dynamic behavior of the tertiary sources. When comparing the measurements with previous results, we assume limited diurnal variations to cover all 17 apartments.

2.5. Statistical analysis

The air concentrations of both data sets were analyzed as PCB_{sum7} with values for single congeners below detection limit given the value zero. Further, air concentrations from data set B were analyzed for different homolog groups. The floor plans are identical with four apartment types and with each apartment type having the same orientation towards compass direction in all buildings. The apartment type (expressed as a categorical variable) is used as one of the explanatory variables in the analysis since the parameters characterizing each apartment, like area, surface area per volume ratio, length of sealant per area etc. are correlated with each other (Fig. 1). In order to test if the real distance between the categories can better explain the observed vales, additional analysis (on univariate level) has been conducted. This included the area (m^2) or surface area per apartment volume (m^2/m^3) or sealant length in the room (kitchen or living room) per room volume (m/m^3), used as scale variable (one at the time).

2.5.1. Data set A

Potential predictors of PCB concentrations were determined by generalized linear models using SPSS (IBM SPSS Statistics 25 for Windows). The residuals of the PCB_{sum7} were not normally distributed neither in the original data nor after natural logarithmic transformation. The negative binomial distribution was chosen and satisfactory goodness of fit was obtained.

Six explanatory variables were investigated (Table 1): average indoor air temperature during measuring period, apartment type, building, season (winter/summer), floor level (≤5, 6–10, ≥11) and year of measurement. In this data set, sampling have been conducted over several years. To account for potential effect of time, the sampling year has been included as variable in the model. All statistical analyses are done separately for kitchen and living room measurements.

The following approaches were used to determine predictors of PCB concentration in indoor air of the contaminated apartments: i) univariate regression analysis, where each explanatory variable was included individually in the model and ii) a multiple regression approach, including all explanatory variables with mutual adjustment and iii) a linear regression with backwards stepwise elimination. The significance threshold was $p \leq 0.05$.

The incidence rate ratio (IRR) expressed as the percentage change (%) in PCB levels in indoor air according to either an increment change in linear predictors or class variable in relation to the reference were calculated from exponentials of the regression estimates according to

$$\boldsymbol{IRR} = (\boldsymbol{exp}(\boldsymbol{estimate}) - 1)\cdot 100\% \qquad \text{Equation 1}$$

2.5.2. Data set B and building characteristics

Multivariate analysis were performed with respect to air concentrations and building characteristics (as Data set A). In total, 51 apartments provided data for the analysis.

Residuals of PCB_{sum7} were tested for normal distribution by use of Q-Q models. As the residuals were normally distributed, generalized linear

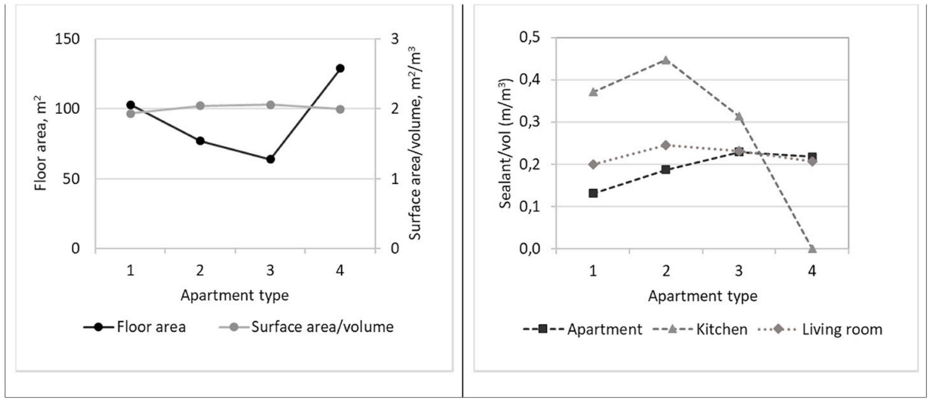

Fig. 1. Parameters characterizing each apartment.

Table 1
Building characteristics analyzed in relation to air concentrations.

Parameter	Apartment type	Floor level	Building	Time (year)	Summer/winter	Temperature
Data set A	x	x	x	x	x	x
Data set B	x	x	x	–	–	x

models (IBM SPSS Statistics 26 for Windows) were performed for the multivariate analysis as well as the univariate linear regression analysis, where each explanatory variable was included individually in the model. Significance of predictors were determined by use of linear regression with backwards stepwise elimination. The significance threshold was $p < 0.05$.

Similar to Data set A, three explanatory variables were investigated (Table 1): apartment type, building and floor level (≤5, 6–10, ≥11).

2.5.3. Data set B and questionnaire

The following behavior questions were used: frequency of vacuuming, dusting, floor washing and airing. The categories of frequency appears from Figs. 3 and 4. In total 52 questionnaires were used (one residence did not provide questionnaire, nor vacuum cleaner bag). In total two residences did not provide a vacuum cleaner bag, i.e. $n = 51$ comparing PCB in dust with cleaning and airing frequencies.

Test for homogeneity was initially used for testing variances between apartments with different cleaning frequencies regarding the three cleaning variables. As there were no significance between the variances, Anova, Least Significant Difference (LSD) test was used to analyze mean PCB contents in apartments with different cleaning frequencies.

3. Results

The results are divided in three sections: i) Analysis related to building characteristics, (Data set A and B), ii) repeated measurements over time (Data set A and B), and iii) analysis of cleaning behavior and airing based on the questionnaires (Data set B).

3.1. Building characteristics

Table 1 shows the parameters tested for the two data sets. An additional analysis (on univariate level) was made on Data set A including the area (m^2).

3.1.1. Data set A

Table S3 shows the characteristic of the apartments included in analysis and the number of observations for the parameters tested. As described before, in some cases, the type 4 apartment has been divided into two. There were only 5 of those apartments investigated and they were excluded from the final regression analysis due to small sample size. The investigated apartments were distributed across all the five contaminated buildings, however with unequal number of measurements. Only four apartments were located in building 1 and six apartments in building 5, while there were over 30 apartments from each of the remaining three buildings. Similar to the apartment type, initial analysis was done on all buildings, but building 1 and 5 were excluded from the final regression analysis.

Most of the measurements were done in summer period (Table S3). The median indoor air temperature (and 5th - 95th percentile) during the sampling periods within the investigation was 23.4 °C (18.6–26.9 °C) in the living room and 23.6 °C (19.2–27.1 °C) in the kitchen. Mean and median values of temperature in different seasons are presented in Table 2. PCB-28 and PCB-52 were found in 100% of living room samples and 98% of kitchen samples and PCB-101 was found in respectively 96% and 95% of the samples. Those three congeners constitute the vast majority of PCB_{sum7}, as the remaining congeners were both less frequent and had lower concentration. PCB-118 was found in approx. 40% of samples, while the remaining three congeners were found in 2–17% of the samples.

The median and mean concentration of PCB_{sum7} for kitchen and living room are shown in Table 3. Table 2 presents the mean and median values of temperatures and air concentrations of PCB_{sum7}, for living room and kitchen, divided by season. The measurements in kitchen and living room were strongly correlated ($R^2 = 0.8$). However, the concentrations measured in the kitchen were significantly higher than the concentrations measured in the living rooms ($p < 0.001$, Wilcoxon signed rank test). When looking at the different apartment types, significant difference between kitchen and living room is only found for apartment type 1 and 2, while for apartment type 3 the difference is just on the significance border ($p = 0.053$, Wilcoxon signed rank test).

Table 2
Mean and median values of temperatures and air concentrations of PCB_{sum7} in living room and kitchen, divided by season.

		Temperature, °C		PCB_{sum7}, ng/m³	
		mean	median	mean	median
Summer	Living room	24.3	24.2	298	258
	Kitchen	24.6	24.5	358	292
Winter	Living room	21.1	21.3	312	289
	Kitchen	21.0	20.8	420	326

Table 3
Mean and median concentration of PCB_{sum7} in indoor air [ng/m^3] measured in living room and kitchen of the investigated apartments. $P < 0.001$ for kitchen-living room comparison.

	N (>LOQ)	Median (5th –95th percentile)	Mean (SD)
PCB_{sum7} – living room	114 (114)	260 (58–682)	301 (192)
PCB_{sum7} – kitchen	110 (108)	310 (84–948)	378 (248)

Taking season into account, apartment type 3 becomes significant in wintertime. For apartment type 4, there is not significant difference between concentration of PCB_{sum7} for kitchen and living room ($p = 0.7$, Wilcoxon signed rank test).

Table 4 and Table 5 show results of univariate-, multivariate- and backward stepwise linear regression analysis for the PCB_{sum7} concentration in air in respectively living room and kitchen.

Among the explanatory variables tested, temperature and season have significant influence on the PCB concentration in the air. Higher indoor air temperature and wintertime are associated with the higher concentrations of PCB in the air. The estimates of multivariate models are mostly in agreement with the univariate model, as for direction, magnitude and significance, except for season, which is not significant in the univariate models but becomes significant in the multivariate model (living room) or multivariate model - backward elimination (kitchen). The same applies for temperature, but only in the kitchen data.

For the measurements conducted in kitchen only, type of the apartment is significant in all three models, with apartment type 4 having lower concentrations than the three other types. When dividing the data by the seasons, the significance is only achieved in the wintertime. As discussed below, the lower concentrations of PCB in apartment type 4 in wintertime can be due to constructional differences. This result should however be viewed with caution due to small sample size (6–9 apartments in each type had measurements in wintertime).

Additional univariate analysis conducted on apartment-characteristic explanatory variables i.e. area (m^2), surface area per apartment volume (m^2/m^3) or sealant length per room volume (m/m^3) does not change the result considerably. Significant correlation has been found between concentration of PCB in the kitchen and each of the apartment-characteristic explanatory variables. Likewise for the apartment type, when dividing the data by the seasons, the significance is only achieved in the wintertime.

3.1.2. Data set B

Number of air samples in different categories of building characteristics are shown in Table S4. Details on air concentrations are presented in Andersen et al. [9]. The indoor temperature (median and 5th - 95th percentile) during the sampling periods within the investigation covering Data set B was 22.0 °C (19.4–24.3 °C) in the living room.

Results of linear regressions with building characteristics and indoor air temperature showed that the temperature was the only parameter being univariate and multivariate significant positive related to the indoor air PCB_{sum7} concentration (Table 6).

3.2. Time

Changes of air concentrations in time were analyzed in two ways: i) all data obtained within investigation A to see, whether these generally show a change during the time span of the collection period (years), ii) the repeated measurements taken within the same apartment, i.e. a change in the single apartment over time.

3.2.1. Data set A

Looking at Data set A and air concentrations vs. time, no relations are seen (Figs. S3 and S4). Neither shows the models significance in any of the analysis (Tables 4 and 5).

3.2.2. Repeated measurements over time

In total 31 apartments were visited with repeating measurements within the period 2011 to late 2017 (see above). 14 apartments were visited twice within Data set A (both living room and kitchen), whereas 17 apartments had a first measurement collected in Data set A and repeated in Data set B (only living room). Fig. 2 compare the oldest to the latest measurement in the living rooms. In general, the data shows some variation over time, though no clear trend was observed.

As the statistics of Data set A show a seasonal variation with higher concentrations in winter time, the repeated measurements from the living rooms were also divided according to season (Fig. S5). Further, the relative difference in air concentration have been compared to the duration of time in between the measurements as well as the concentration of the starting point (oldest measurement) (Fig. S6). Neither season, nor duration of time between samples seem to explain the differences. Although with a big scatter, the lower starting points tend to have the highest relative increase (Fig. S6b).

3.3. Questionnaire

The air and dust concentrations are related to self-reported frequencies of airing and different cleaning parameters below.

3.3.1. The impact of airing and cleaning frequencies on air and dust concentrations

Fig. 3 shows the average and standard deviation (±SD) of air (PCB_{sum7}) and dust (PCB_{sum15}) divided in the categories of frequency of airing. The results related to airing do not show any tendencies and gave no rise to further statistical analysis.

Fig. 4 shows the average and standard deviation (±SD) of air (PCB_{sum7}) and dust (PCB_{sum15}) concentrations, divided in the categories of frequency of vacuuming, dusting and floor washing.

The statistical analysis of the individual cleaning methods compare all categories of frequency with each other, i.e. not necessarily the increasing time of between cleaning. In general, the PCB concentration in air and dust was significantly or near significantly higher in apartments where the cleaning frequency was seldom (one or twice a month) compared to apartments with more frequent cleaning (Fig. 4 and Table S5). This tendency was not observed for floor washing and concentration of PCB in dust, where only washing floors twice a month was significantly higher than monthly. Vacuuming twice a month was

Table 4
Univariate and multivariate association between the concentration of PCB_{sum7} in indoor air of living room and available explanatory variables.

	Univariate association		Multivariate association		Multivariate model - backward elimination	
	Incidence rate ratio, % (95% CI)	p-value	Incidence rate ratio, % (95% CI)	p-value	Incidence rate ratio, % (95% CI)	p-value
Apartment type	−0.6 (−12; 12)	0.92	0.0 (−11; 13)	0.99		
Season	3.6 (−23; 39)	0.81	52 (5.5; 119)	0.02	48 (4.8; 108)	0.03
Temperature	7.7 (2.0; 14)	0.01	13 (5.8; 20)	<0.01	12 (5.4; 20)	<0.01
Time/year	4.0 (−5.2; 14)	0.41	5.7 (−3.7; 16)	0.24		
Building	9.8 (−6.9; 29)	0.26	11 (−6.7; 33)	0.23		
Floor level	−5.8 (22; 13)	0.53	−5.4 (−21; 14)	0.56		

Table 5
Univariate and multivariate association between the concentration of PCB_{sum7} in indoor air of kitchen and available explanatory variables.

	Univariate association		Multivariate association		Multivariate model - backward elimination	
	Incidence rate ratio, % (95% CI)	p-value	Incidence rate ratio, % (95% CI)	p-value	Incidence rate ratio, % (95% CI)	p-value
Apartment type	−17 (−27; −5.0)	0.01	−20 (−30; −7.7)	<0.01	−19 (−29; −7.0)	<0.01
Season	16 (−16; 64)	0.37	46 (−4.1; 121)	0.08	51 (0.9; 125)	0.05
Temperature	3.2 (−3.0; 9.8)	0.32	11 (2.7; 19)	0.01	10 (2.2; 19)	0.01
Time/year	2.6 (−7.4; 14)	0.62	6.7 (−3.9; 19)	0.23		
Building	3.4 (−14; 25))	0.73	2.4 (−16; 24)	0.81		
Floor level	−5.9 (−24; 16)	0.57	−2.3 (−20; 20)	0.82		

Table 6
Building characteristics and air concentrations (PCB_{sum7}), n = 49.

	Univariate association		Multivariate association		Multivariate backward elimination	
	Incidence rate ratio (95% CI)	p-value	Incidence rate ratio (95% CI)	p-value	Incidence rate ratio (95% CI)	p-value
Apartment type	−41.2 (−93.6; 11.2)	0.121	−17.9 (−60.8; 24.9)	0.402		
Temperature	50.9 (22.1; 79.6)	0.001	50.8 (20.7; 78.5)	0.001	51.4 (22.9; 79.8)	0.001
Building	0.46 (−29.1; 29.9)	0.975	21.5 (−50.6; 35.9)	0.734		
Floor level	28.0 (−35.5; 91.6)	0.379	28.6 (−30.9; 84.3)	0.356		

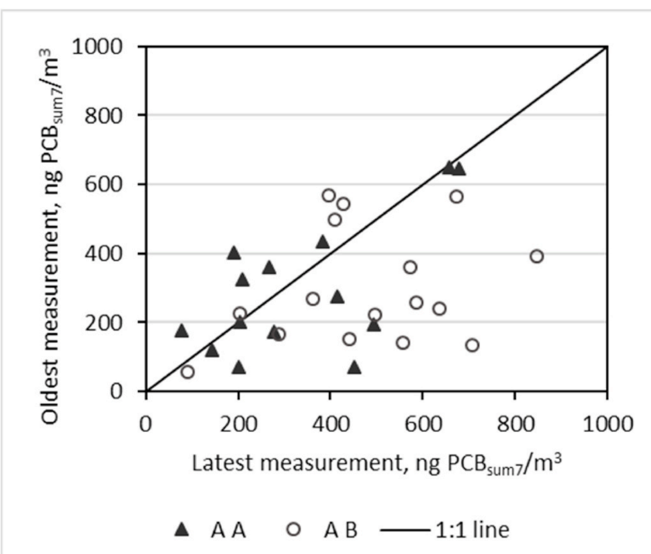

Fig. 2. Repeated measurements of air concentrations over time in the same apartment (living room) within the period 2011 to late 2017. The oldest vs. the latest measurement for data obtained within investigation A "A A" and investigation A measurements repeated in investigation B "A B".

related to higher air concentrations than vacuuming daily or weekly. Floor washing twice a month or dusting monthly were associated with higher air concentrations than all the other groups. The sample size of the category "monthly" were only 3–6 observations. For dust concentration of PCB_{sum15} (Fig. 4 right) vacuuming twice a month showed higher levels than daily or twice a week, while monthly dusting showed higher values than all other groups. The same differences were seen analyzing the concentration of PCB_{sum7} in dust (Table S5). Comparing air and dust for the individual cleaning methods, it is the same category of cleaning frequencies showing significant differences to some or all other groups (Fig. 4).

3.3.1.1. Homolog groups. Looking at the homolog groups, the air concentrations were significantly higher for homolog groups 4 Cl and 5 Cl in apartments with a floor washing twice a month compared to higher frequencies of floor washing (p = 0.005 and p = 0.020, respectively). Concentration of the homolog groups 2 + 3 Cl, 4 Cl, 5 Cl and 6 + 7 Cl in dust in relation to cleaning frequencies are shown in Fig. S7. Significantly higher concentration of homolog group 2 + 3, 4 and 5 in dust were seen in apartments with a dusting frequency of one a month compared to higher dusting (p = 0.007, p = 0.003 and p = 0.005, respectively).

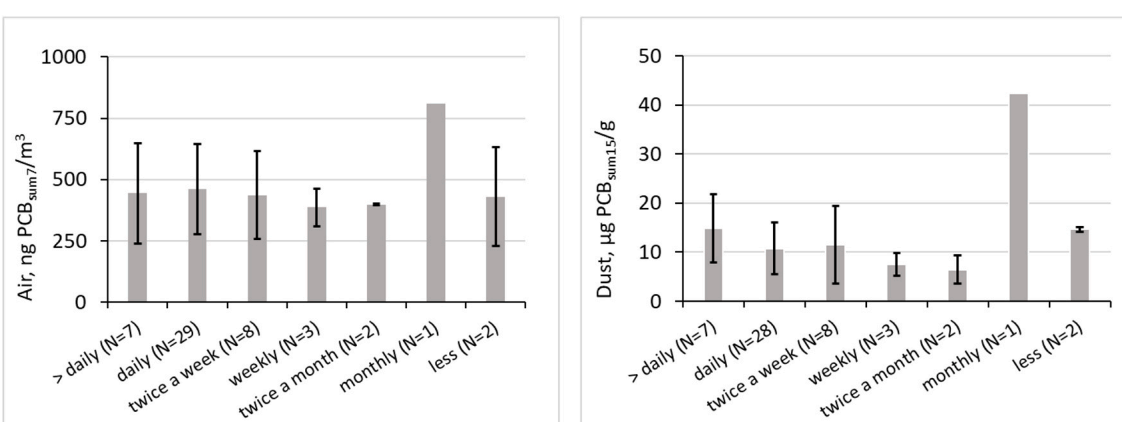

Fig. 3. Mean values (±SD) of air and dust concentrations, divided in the categories of frequency of airing. (Airing more than one time a day: ">daily").

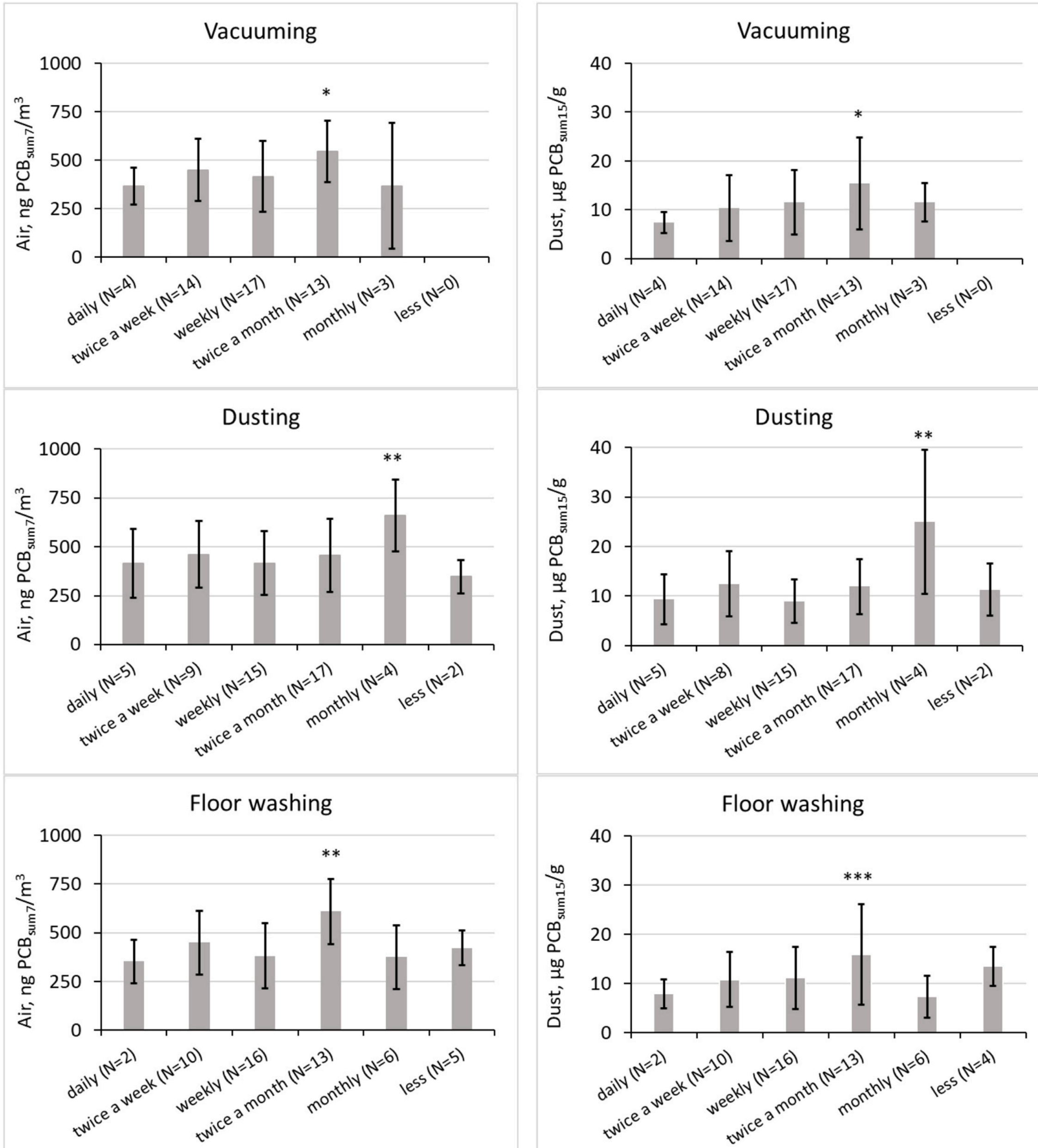

Fig. 4. Mean values (±SD) of air concentrations (left) and dust concentrations (right), divided in the categories of frequency of vacuuming, dusting and floor washing. * different from daily and weekly (air) or twice a week (dust), ** different from all other categories, *** different from monthly.

4. Discussion

4.1. Air concentrations, temperature and building characteristics

The air concentrations in the apartments can be influenced by many factors as also discussed in Andersen et al. [9]. The relation to temperature found in data set A is in line with Data set B [9]. Data set A, providing data for both kitchen and living room as well as different seasons, show higher air concentrations of PCB associated with higher temperature and wintertime. As mentioned in the introduction, higher air concentrations related to higher temperature has been reported, also in investigation B. In this case, dividing Data set A in seasons, differences for the measurements conducted in the kitchens are revealed. The type of apartment is significant, with type 4 having lower concentrations than the other types, though only in wintertime. We interpret this as a consequence of constructional differences as the kitchen in apartment type 4 was facing a concrete façade, whereas the other apartments have kitchens (and balconies) in the light facade with indoor PCB containing sealants. We speculate that the heating pipes below the floor have an impact on the emission strength of the sealants situated here. This is in good compliance with increased air concentrations in wintertime associated with the heating demand, having an impact in kitchens facing the light façade, but not the apartment type 4. Data set A contain air concentrations in both kitchen and living room, showing good correlation, though in apartment type 1 and 2 (and type 3 in wintertime) the air concentrations in the kitchen were significantly higher than the living room. Both kitchen and living rooms in these apartment types are facing the light façade having sealants, though length of sealant relative to volume of the room are slightly higher in the kitchens than in living rooms in type 1 and 2 (Fig. 1). Further, mechanical exhaust ventilation is placed in the kitchen. The slightly higher source strength, expressed as length of sealant relative to volume of room, together with the exhaust ventilation in the kitchen could explain these results. It is further supported by the observation, that no difference is seen between living room and kitchen in apartment type 4, where the kitchen does not have indoor sealants and draw in air from other room with sources.

Apart from indoor temperature and apartment type, we did not find clear relations to parameters giving an explanatory framework for the levels and thereby a possibility to predict levels in apartments where no measurements have been done. The building characteristics were chosen to test different hypothesis of influencing factors having impact on the air concentrations. Whereas the floor area of the apartment types was different, the surface area to volume ratios were very similar. Further, the sealant length to volume ratio showed no clear difference for the apartment types with exception of kitchen in the apartment type 4 (Fig. 1). This lack of differences may be the reason for e.g. different source strength is not reflected. The apartment type might reveal impact from compass direction and with that different degree of influence from sun and heating in summertime. The turbulence and wind speed around the high-rises and the indoor upward pull of air might have an influence on air exchange rates and the degree of outdoor air and internal transfer of air between apartments and floors. We have analyzed this by dividing the data into groups of floor levels. The buildings were tested to look for changes occurring during the construction, i.e. change in PCB product or mixture, though analysis of the composition of congeners in air do indicate the use of the same product in all five contaminated high rises [9]. None of these parameters showed relations in the air concentrations in the analysis.

4.2. Air concentration and time

Air samples taken in the same apartment at different times (Fig. 2) reveal large variations, though no clear trend was observed. One could question whether differences in concentration levels with the first sample associated with investigation A and the second in investigation B, would be a consequence of the differences in sampling procedure or analytical laboratories; however, the variation is also seen within repeated samples of investigation A. We do not expect an impact lowering the air concentrations due to the procedure with airing and conditioning before measure in investigation A. As also mentioned below (section 4.3) we expect the dynamic behavior of the tertiary sources to counteract changes in air concentrations. Here we compared short-term and daily measurements, though earlier observations during conditions with stable indoor temperatures showed limited diurnal variations [9]. Therefore, we believe, that the results express an actual variation in levels rather than procedure. The effect of season or time in between repeated samples does not seem to explain the variations seen for these data. There is a weak tendency of increased relative differences between oldest and latest measure and the concentration level of the starting point. The lower the concentration at starting point, the higher relative difference. The buildings were raised more than 40 years ago and one could speculate whether the apartments have reached a relatively stable air concentration due to saturation of the tertiary sources. Maintenance activities i.e. sanding and varnishing floors, removing wallpaper and adding new paint upon moving in are expected to have an impact on the air concentrations as new absorbing surface materials are introduced to the rooms. A study demonstrated an immediate reduction of air concentrations in PCB contaminated rooms by introducing absorbing materials with considerable surface areas [25]. The sealants are located behind wooden panels or below the floor and therefore, we do not expect maintenance activities to affect these primary or secondary sources. Depending on the amount of activities, we expect a reduction in the air concentrations as the new materials will sorb PCB from the room air. In the following time, air concentrations will increase as the uptake of PCB in the sorbing material increase until a near steady state again is achieved between the materials and air. It is speculated whether some of the variation seen in Fig. 2 could be due to impact from maintenance activities.

4.3. Airing and cleaning habits

The air concentrations (PCB_{sum7}) obtained in Data set B did not show clear relations to airing. At equilibrium conditions, the air concentration will depend on the emission strength of the sources, volume of apartment and the air exchange rate. The influence of changes in air exchange rate can differ significantly as reported by Lyng et al. [15,16] and in this building estate with indoor sealants as PCB source, we would expect that the dynamic behavior of the tertiary sources to a large extent is able to counteract changes in air concentrations. Frequent airing have many advantages for the indoor air quality, though for the air concentrations of PCB we have not been able to see an effect, probably due to counteracting tertiary sources.

Cleaning habits did show a minor effect on air and dust concentrations as more frequent cleaning resulted in significant lower concentrations than less frequent cleaning in all categories, though the variabilities were high and the effect not pronounced. The monthly frequency was not necessarily relating to higher concentrations than a frequency of cleaning twice a month, though monthly frequency also had considerably lower number of observations. A similar trend between air concentrations and cleaning frequencies was observed in another Danish building estate with PCB containing sealants [7] and significance was found for homolog groups with 5 or more chlorine [13]. Comparing concentrations (PCB_{sum7}) in dust and air in BSP, we found a positive correlation, though the variability was high [9]. The establishment of near steady state conditions between the air concentrations and settled house dust can be relatively fast, though it depends on the congener and chlorination degree. Comparing the dust samples from the apartments to experiments with dust-air partitioning, the general picture was dust samples close to steady state [26]. Future statistical analyses will be made looking at the cleaning frequencies and other variables to investigate, whether other factors might have an impact on the concentration levels.

5. Conclusion

The data shows that higher indoor air temperature and wintertime were associated with the higher concentrations of PCB in the air. It is speculated that the higher air concentrations in cold weather are due to radiator pipes heating PCB sources under the floors. No relations were seen between air concentrations and building characteristics such as apartment type, floor level and building. Therefore, we were not able to provide a scheme for gap-filling of exposure data for an epidemiological study of effect of living in PCB-contaminated housing. Repeated measurements of air concentrations measured in a number of apartments during the period showed some variation, though no clear trend. This might relate to maintenance activities, expected to have an impact on the air concentrations. Cleaning habits did show a significant influence on the air and dust concentrations, though the effect was not pronounced. Apparently, airing did not influence the concentration levels.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.

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