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Full length article



## Silicone wristbands as personal passive samplers of exposure to polychlorinated biphenyls in contaminated buildings

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### ABSTRACT

Polychlorinated biphenyls (PCBs) were used in a number of industrial products from 1950 to 80s, including building materials. As a result, some buildings exhibit high levels of PCBs in the indoor environment. The aim of this study was to test silicone wristbands as a method for estimating personal exposure to PCBs in buildings both in controlled experiments and field settings. In the controlled study, the sampling kinetics of silicone wristbands were investigated in a 31-day uptake study. The field study focused on the application of wristbands as a personal exposure measure. It included 71 persons in a contaminated housing estate and 23 persons in a reference group. The linear uptake of PCBs ranged from 2 to 24 days for PCB-8, 18, 28, 31, 40, 44, 49, 52, 66, 99, and 101 under controlled conditions. A generic sampling rate ( $R_k$ ) of  $2.3 \text{ m}^3 \text{ d}^{-1}$  corresponding to a mass transfer coefficient of  $17 \text{ m h}^{-1}$  was found in the controlled kinetic study. Partitioning coefficients were also determined for the nine congeners. In the field study, an apparent generic field sampling rate ( $R_f$ ) of  $2.6 \text{ m}^3 \text{ d}^{-1}$  was found; when adjusted to reported hours exposed, it increased to  $3.5 \text{ m}^3 \text{ d}^{-1}$ . The wristbands were shown to be a good tool for predicting airborne exposure, as there was a highly significant difference between the exposed and reference group as well as a clear trend when used for ranking of exposure. In correlation analyses, highly significant correlations were observed between air and wristband levels, though adjusting by self-reported exposure time only increased the correlation marginally in the field study. The obtained kinetic data can be used for estimating the magnitude of external exposure. The advantages provided by the wristbands in the form of easy use and handling are significant, though the limitations should also be acknowledged.

### 1. Introduction

Polychlorinated biphenyls (PCBs) were used from 1950 to the 1980s in a variety of applications including building materials to which they were added as flame retardants and plasticizers ('open applications') until 1977, and as dielectric fluid in e.g. fluorescent lighting ballasts ('closed applications') until 1986, when they were banned in Denmark due to their persistent, bioaccumulative and toxic (PBT) properties. PCBs were among the first groups of chemicals on the Stockholm Convention that regulates Persistent Organic Pollutants globally and entered into force in 2004. Later, the International Agency for Research on Cancer (IARC) classified all PCB congeners as Group 1 human carcinogens (Lauby-Secretan et al. 2013).

In many cases, the original PCB-containing building materials are still in use. Over the years, PCBs have evaporated from open applications or potentially leaked from closed applications, resulting in contaminated indoor environments and ongoing human exposure. PCBs used in elastic sealants in the 1960s and 1970s can still cause high PCB levels in the indoor environment today (Andersen et al. 2020; Frederiksen et al. 2012), leading to elevated PCB blood levels of the inhabitants, particularly of the lower chlorinated PCB congeners (Frederiksen et al. 2020; Meyer et al. 2013). The health risks associated with living with such high levels of exposure have recently been investigated. A register study indicated an increased risk of cryptorchidism among sons of mothers exposed in these (and similar) buildings during pregnancy (Kofod et al. 2021), while a relatively small clinical study showed only marginal and

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inconsistent associations between prenatal exposure and male reproductive function (Tøttenborg et al. 2022). Further register studies investigating risk of cancer and cardiovascular disease following exposure in these buildings are currently under way.

Indoor air concentrations – and deduced exposure estimates – of PCBs are typically determined by active sampling, i.e. passing a defined volume of air through a sorbent material. An alternative is the uptake of chemicals in the gas phase on passive samplers, which can either be described by a one-box model or a mass transfer model; the relation between parameters of these are described by Bartkow et al. (2005). The uptake is characterized by three stages: the linear uptake phase (also called the kinetic phase); the curvilinear phase; and the equilibrium phase. It is often desirable to sample in the linear phase, where the surface resistance is negligible and uptake rates are at their maximum (Shoeib and Harner 2002). Alternatively, sampling in the equilibrium phase is warranted, to be able to apply equilibrium partition coefficients, while the curvilinear phase is more challenging to describe (Mayer et al. 2003).

Silicone (polydimethylsiloxane, PDMS) samples non-polar, semi-volatile organic compounds (SVOCs) such as PCBs very efficiently. The capacity per volume of sampler is markedly higher for silicone than for polyurethane foam (PUF)-samplers resulting in a longer linear uptake phase (Tromp et al. 2019). While silicone has been used for passive sampling of non-polar, organic contaminants in the aquatic environment for some time (Booij et al. 2016), it has only recently been applied in human exposure monitoring e.g. for indoor air sampling or more commonly as wristbands or lapels for personal sampling (O'Connell et al., 2014; Okeme et al. 2018; Vorkamp et al. 2016). The format of the wristband has the advantages of being a cheap and easy-to-apply tool, which does not require special sampling equipment and can be sent by mail. Studies have shown that SVOCs are distributed homogeneously in the sampler (Levasseur et al. 2022) and the wristbands can be stored for at least 6 months after sampling without degradation or loss of a wide range of SVOCs including PCBs (Anderson et al. 2017). However, the wristbands also have some limitations as they sample from air but potentially also from surfaces and skin contact, and the uptake can be altered by coverage of clothes, making it difficult to disentangle exposure components. Thus they are best suited for semi-quantitative exposure estimates. The strengths and limitations of applying silicone wristbands for personal exposure estimate has been extensively discussed in a recent review (Samon et al. 2022).

Since their introduction into exposure assessment, silicone wristbands have been used to measure exposure to some volatile organic compounds (VOCs) and several classes of SVOCs, including brominated flame retardants (BFRs), organophosphate esters (OPEs), plasticizers, pesticides, phenols, and polycyclic aromatic hydrocarbons (PAHs) (Dixon et al. 2019; Hamzai et al., 2021). They have been used both qualitatively by differentiating exposure profiles (Aerts et al. 2018) as well as quantitatively for ranking exposure. Wristbands have also been shown to be predictors of personal exposure to OPEs, some BFRs, PAHs, nicotine, phthalates, and environmental phenols through positive associations with human biomarkers, many of which were comparable to or better than hand wipes, house dust, and active air samplers (Dixon et al. 2018; Hammel et al. 2020; Hammel et al. 2016; Hammel et al. 2018; Levasseur et al. 2021; Quintana et al. 2019). To date, several studies have also evaluated personal exposure to PCBs using wristbands among office workers (Young et al. 2021), children (Travis et al. 2020), and adults from the general population (Dixon et al. 2019; Doherty et al. 2020; Wise et al. 2020). However, most have reported extremely low PCB concentrations and detection frequencies for the analyzed PCB congeners.

The current study was conducted in a social housing estate near Copenhagen, Denmark. The estate consists of 12 identical high-rise buildings surrounded by many lower apartment buildings. At the time of construction (1969–74), PCB-containing sealants were used in the construction of the first five high-rise buildings, whereas non-PCB-

sealants were used in the remaining seven high-rises. The five high-rises are heavily contaminated with PCB (median air level: 2164 ng PCB<sub>total</sub>/m<sup>3</sup>), while the remaining seven have approximately 50 times lower PCB-levels (median air level: 49 ng PCB<sub>total</sub>/m<sup>3</sup>) (Andersen et al. 2020). PCB<sub>total</sub> is the parameter used in Danish recommended indoor air action values and is calculated as: PCB<sub>total</sub> = 5 × Σ(PCB-28, 52, 101, 118, 138, 153, 180); the lowest Danish action value for indoor air is 300 ng/m<sup>3</sup> and the upper is 3000 ng/m<sup>3</sup> (see further in Andersen et al. (2020)). PCB<sub>tot</sub> is often used for regulatory purposes in Europe. The contaminated high-rises have provided the settings for multiple studies on e.g. the dynamics of PCBs in indoor environments (Andersen and Frederiksen 2021; Lyng et al. 2016a; Lyng et al. 2016b; Morrison et al. 2018), exposure characterization (Andersen et al. 2020; Frederiksen et al. 2020) as well as clinical and epidemiological studies (Kofod et al. 2021; Tøttenborg et al. 2022). In the current study, silicone wristbands were used as an exposure measure as part of a larger study including levels of PCBs in indoor air, dust and surfaces wipes as well as samples of serum and hand wipes of residents in both contaminated and uncontaminated parts of the estate (Andersen et al. 2020; Frederiksen et al. 2020). Shortly after the sampling for this project, all residents of the five contaminated high-rise building moved out and the buildings are currently being demolished.

The aim of this study was to evaluate the utility of silicone wristbands as a method of personal sampling for determining PCB exposure in buildings in controlled experiments and under field conditions. The study included investigations into the kinetic uptake of PCBs in the wristbands under controlled conditions and field applications (i.e., building residents wearing wristbands) for personal exposure estimates in PCB-contaminated apartments.

## 2. Materials & methods

The study included two sets of experiments, both of which took place in the high-rise buildings: a “kinetics study”, which focused on uptake of PCBs over time, and a “field study”, during which 71 residents from the contaminated and 23 residents from the reference sections wore wristbands. The distribution between exposed and reference group was determined in the main study of blood and air and was a compromise of budget and getting the best possible description of the exposed group with some references. The kinetic study was performed in a vacated apartment in one of the contaminated high-rises under controlled conditions, with wristbands collected for a time series of up to 31 days. For the participants in the field study, a clear exposure contrast had been found between the exposed and reference groups in all other matrices, as described in detail elsewhere (Andersen et al. 2020; Frederiksen et al. 2020).

### 2.1. Pre-treatment of wristbands

Impurities of the silicone are co-extracted with the PCBs in chemical analysis, causing increasing baseline noise and interfering peaks as well as resulting in loss of sensitivity in the analysis. To prevent this, the silicone wristbands (202 × 12 × 2 mm, Nordic Wristbands, Denmark) were pre-cleaned prior to deployment by heating them to 280°C for 5 h. Shorter heating time resulted in unacceptable interferences in the instrumental analysis, while longer heating resulted in brittle wristbands that were not well suited for field studies. Heating resulted in an approx. 5% weight loss of the wristbands, and the density of the cleaned wristbands were 1.08 g/cm<sup>3</sup>. The wristbands were stored in sealed Rilsan bags at room temperature until deployment.

### 2.2. Kinetics study

Since it is desirable to collect passive samplers while they are sampling linearly (kinetic phase) (Shoeib and Harner 2002), the kinetics of the wristbands were investigated in a 31-day experiment carried out in a

vacated apartment in one of the high-rise buildings also hosting the field study. The experiments were carried out in October and November and the mean outdoor temperature was 6.6 °C in the measurement period (DMI, 2022); all windows and doors were closed, only the research group and janitor had access to the apartment. The apartment had a well-described, high and stable PCB-level in the air (~4000 ng PCB<sub>total</sub>/m<sup>3</sup>) and had been used in a number of other experiments with PCBs, e.g. by Lyng et al. (2016a). Pre-cleaned wristbands were hung on a horizontal stainless steel rod approx. 40 cm above the floor; air movement was ensured by a fan placed approximately 2 m from the wristbands blowing softly across the room. The temperature was logged every 10 min using a HOBO-U12-012 data logger (Onset, Bourne, MA, US); the 31-day temperature average was 21.0 ± 0.36 °C. Wristbands were taken down on day 3, 5, 6, 7, 8, 9, 10, 12, 14 and 31. Altogether 18 wristbands were collected for the time series, two per time point except at day 3 and 31 where only one band was collected. In addition, five extra wristbands were collected on day 7 for validation purposes, and a field blank was collected at the beginning of the experiment, t<sub>0</sub>. The wristbands were stored in individual Rilsan bags at -20°C until extraction and analysis. Due to the stable conditions, only a single 24 h active air sample was taken for calibration, which was done on day 9. The sampling and analysis of this sample followed the methodology described in Andersen et al. (2020), including logging of room temperature (24 h average: 21.3 ± 0.03 °C).

### 2.3. Field study: exposure study of residents in contaminated buildings

Pre-cleaned silicone wristbands were given to 71 residents of PCB-contaminated high-rise apartment buildings and 23 participants in a reference group from the same housing area at the time of blood sampling (see Frederiksen et al., 2020). The participants were instructed to wear it at all times for 7 days, to place it in a Rilsan bag at the end of the sampling period, seal the bag and deliver it to a dedicated mailbox located in each high-rise. The wristbands were collected from the mailboxes within 24 h upon return and stored at -20 °C until analysis. During the 7 days, the participants filled in a simple questionnaire on the hours spent in the apartment per day. Furthermore, 5 participants were given one extra wristband, which they were instructed to leave on a horizontal surface in their living room for the same 7 days as they wore a wristband. The interest of participation was high, which enabled sampling from almost all participants though some were challenged by the size of the wristband. The field study was approved by the Regional Ethics Committee (H-16041946).

### 2.4. Chemical and data analysis

Approximately 1 cm (0.3 g) of each wristband was cut off for analysis. It was cut into smaller pieces, weighed and transferred to a 5 ml stainless steel cell pre-filled with approximately 2 ml of Hydromatrix®, and <sup>13</sup>C-labelled standards (Wellington Laboratories, Canada) were added. The extractions were carried out by pressurized liquid extraction (Dionex, ASE-300) with n-hexane:acetone (1:1), static time 10 min at 125 °C, 4 cycles, 60 % flush and 60 sec. purge. The extracts were concentrated to approximately 1 ml under a gentle stream of nitrogen. The extracts were analyzed by GC-MS-MS (Bruker Scion TQ; column: 30 m SGE HT-8; id: 0.25 mm; film: 0.25 μm) for PCB-8, 18, 28, 31, 40, 44, 49, 52, 66, 99, 101, 105, 118, 138, 153, and 180 (Sigma-Aldrich, Germany) by isotope dilution. <sup>13</sup>C-labelled internal standards were not available for all isomers, and the nearest homologue was used: <sup>13</sup>C-PCB-28 (for 8–31), 52 (for 40–66), 101 (for 99–105), 118 (118), 138 (138), 153 (153), 180 (180). The temperature program started at 80 °C for 2 min, 30 °C/min to 170 °C, 3 °C/min to 300 °C, which was held for 5 min, with a final ramp of 20 °C/min to 325 °C. The limit of quantification (LOQ) was set to a signal-to-noise ratio of 10 in the individual samples, corresponding to approximately 0.01–0.1 ng per sample for each congener. Levels below LOQ were set to zero in the further data

treatment.

The study included a number of blank samples. In addition to procedural laboratory blanks (n = 5), ‘field blanks’ (i.e. pre-cleaned wristbands brought and repacked at all locations where wristbands were handed out) (n = 4) and ‘mailbox blanks’, which were stored in sealed Rilsan bags in the mailbox until pick-up in the contaminated high-rise apartment buildings (n = 8), were also investigated. The levels of laboratory and mailbox blanks were generally below the detection limit, while very low levels of lower chlorinated PCBs were detected in the field blanks, however only at levels around LOQ and well below all samples. Further details can be found in the [supplementary material](#).

All curve fitting and statistical analyses were done in GraphPad Prism version 8.0.2 for Windows (GraphPad Software, San Diego, California USA, <https://www.graphpad.com>). Non-parametric tests, including Spearman Ranks, Mann-Whitney, and Kruskal-Wallis tests, were applied as the data was not normally distributed. In the Spearman Rank correlations non-detects were included in the model, as the assigned rank is independent of the imputed level, as long as these samples are the lowest. Excluding non-detects would lead to a biased model as these samples are truly low.

### 2.5. Calculations

The uptake rates and equilibrium concentrations were modelled by the equation:

$$C = C_{\infty} \cdot (1 - e^{-kt}) \quad (1)$$

where C is the concentration [ng/g] in the wristband at a given time, t [d]. C<sub>∞</sub> [ng/g] is the concentration at equilibrium and k is the rate constant [d<sup>-1</sup>].

The time it takes the wristband to reach equilibrium was evaluated as the time, t<sub>95</sub>, which it takes to reach 95% of C<sub>∞</sub>:

$$t_{95} = \frac{-\ln 0.05}{k} \quad (2)$$

Shoeb and Harner (2002) arbitrarily defined t<sub>25</sub> as a the upper bound of the kinetic phase; in the current study, it was used as an indicator for the length of the kinetic phase. t<sub>25</sub> can be calculated as:

$$t_{25} = \frac{-\ln 0.75}{k}$$

The passive samplers (in the kinetic range) can be calibrated against the results of an active air sample, as described by Vorkamp et al. (2016), to determine the sampling rate of the passive sampler:

$$R = \frac{n_{ps}}{C_{air} \cdot \Delta t} \quad (4)$$

where R is the sampling rate [m<sup>3</sup>/d]; n<sub>ps</sub> is the amount of PCB in the passive sampler [ng]; C<sub>air</sub> is the PCB in air measured by active sampling [ng/m<sup>3</sup>]; and Δt is duration of sampling [d]. A constant air concentration as was assumed.

Sampling of SVOCs (with K<sub>oa</sub> > 10<sup>7</sup>) like PCBs on a suitable passive sampler medium is air-side controlled in the linear phase, thus the mass transfer coefficient (MTC), also called the dry gaseous deposition velocity, can be approximated to the air-side mass transfer coefficient, k<sub>a</sub> [m/d] (Shoeb and Harner 2002):

$$MTC \approx k_a = \frac{R}{A_{ps}} \quad (5)$$

where A<sub>ps</sub> is the surface area of the sampler [m<sup>2</sup>]. Sampling from both inner and outer surfaces of the wristbands was assumed.

The application of a generic sampling rate across a wide ranges SVOCs is supported by the finding of similar sampling rates of 155 compounds across eight chemical groups described by Tromp et al. (2019). In the current study, the sampling rate and MTC was calculated for each congener on each wristband separately, but for the evaluation

of the generic sampling rate for the different conditions (e.g. ‘kinetic study’, ‘field study-active’, ‘field study –static’), the mean of each wristband was used in the further data treatment.

### 3. Results

#### 3.1. Kinetics study

All analyzed congeners were detected above LOQ in all samples from the kinetic study except PCB-180, which had a detection frequency of 33%. Generally, there was good agreement between replicates, and there was a clear increasing trend in concentration with increasing exposure time (Fig. 1). At day 7, five extra wristbands were sampled to estimate the reproducibility. A summary of QA/QC parameters for the method can be found in the [supplementary material](#).

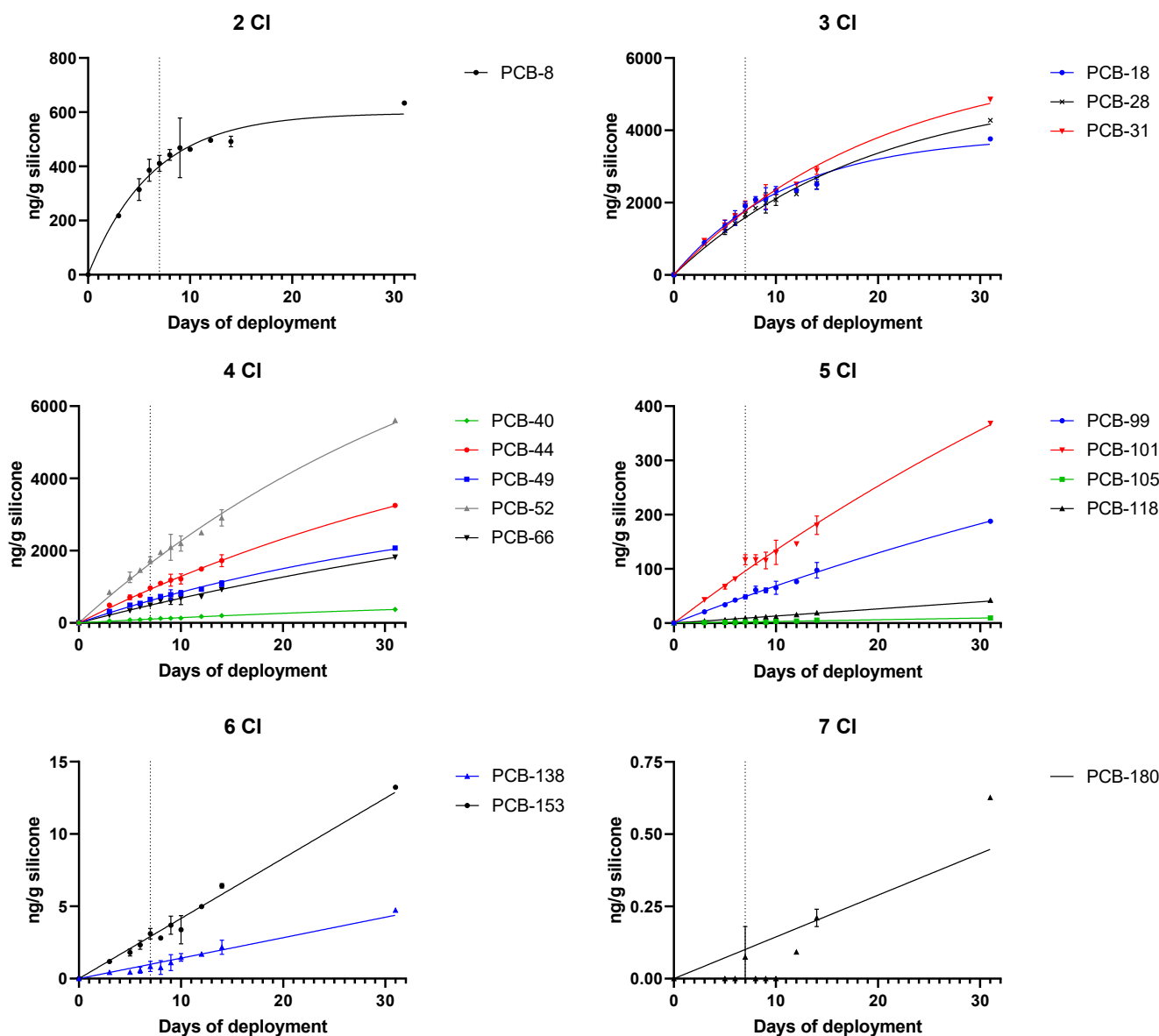
In the initial phase, the uptake is at the maximum, and the sampling rate was calculated using Eq. (4) and data from day 7 of the experiment (Table 1). There was some variation between congeners but no clear trend could be observed (Table S2), and the generic sampling rate was

**Table 1**

Sampling kinetics for silicone wristbands at  $21.0 \pm 0.36$  °C. k: rate constant;  $t_{25}$ : estimated duration of the linear phase;  $t_{95}$ : time-to-equilibrium; and  $C_{\infty}$ : estimated equilibrium concentration in silicone.

Congener	R <sup>2</sup> of fit	k [d <sup>-1</sup> ]	t <sub>25</sub> [d]	t <sub>95</sub> [d]	C <sub>∞</sub> [ng/g]
PCB-8	0.93	0.1	1.8	19	$6.0 \times 10^2$
PCB-18	0.96	0.088	3.3	34	$3.9 \times 10^3$
PCB-28	0.98	0.052	5.6	58	$5.2 \times 10^3$
PCB-31	0.98	0.049	5.8	61	$6.1 \times 10^3$
PCB-40	0.98	0.020	15	154	$8.2 \times 10^2$
PCB-44	0.99	0.021	14	141	$6.7 \times 10^3$
PCB-49	0.98	0.027	11	113	$3.6 \times 10^3$
PCB-52	0.98	0.025	12	121	$1.0 \times 10^4$
PCB-66	0.97	0.016	18	189	$4.7 \times 10^3$
PCB-99	0.98	0.012	23	244	$6.0 \times 10^2$
PCB-101	0.97	0.013	22	227	$1.1 \times 10^3$

2.3 m<sup>3</sup>/d, which corresponds to an MTC of 17 m/h (Table 2) based on the congeners PCB-8 to PCB-101. For these congeners, the partitioning coefficient between silicone and air was estimated (Table 3) based on



**Fig. 1.** PCBs in wristbands deployed 3 to 31 days in a well-characterized, contaminated indoor environment. Day 3 and 31 are single samples, and the remaining samples are duplicates. Mean and standard deviation are displayed and fitted to an exponential plateau function (Eq. (1)). The dotted line is day 7, which corresponds to the deployment time in the field study.



**Table 2**

Overview of mean generic sampling rates (range) of sampling categories across wristbands for congeners from PCB-8 to 101. Field rates: based on exposed group only. Static: based on wristbands left on a horizontal surface in contaminated apartments.

	Sampling rate, whole wristband [m <sup>3</sup> d <sup>-1</sup> ]	Sampling rate, area specific [m <sup>3</sup> d <sup>-1</sup> dm <sup>-2</sup> ]	Mass Transfer Coefficient [m h <sup>-1</sup> ]	Relative Standard deviation (RSD)
Kinetics study (day 7) (n = 7)	2.3 (2.0–2.9)	4.1 (3.6–5.1)	17 (15–21)	13%
Field study –active, apparent (n = 64)	2.6 (0.39–7.3)	4.6 (0.69–13)	19 (2.9–54)	54%
Field study –active, adjusted for home-time. (n = 50)	3.5 (1.1–10)	6.2 (2.0–18)	26 (8.2–75)	57%
Field study –static. (n = 5)	0.57 (0.30–1.3)	1.0 (0.52–2.3)	4.2 (2.2–9.4)	64%

**Table 3**

Overview of estimated partitioning coefficient between silicone and air, note different units (density of wristbands in this study: 1.08 g/cm<sup>3</sup>).

Congener	This study log(C <sub>∞</sub> /C <sub>air</sub> ) [m <sup>3</sup> /m <sup>3</sup> ]	Tromp et al. (2019), log K <sub>sa</sub> [m <sup>3</sup> /m <sup>3</sup> ]	Gilbert et al. (2016), log(C <sub>∞</sub> /C <sub>air</sub> ) [L/kg]	Vorkamp et al. (2016) <sup>a</sup> , log(C <sub>∞</sub> /C <sub>air</sub> ) [L/kg]
PCB-8	6.8			
PCB-18	6.9			
PCB-28	7.2	7.13	7.11	6.4 / 5.8
PCB-31	7.0			
PCB-44	7.4		7.68	
PCB-52	7.4	7.44	7.56	6.5 / 6.3
PCB-66	7.6			
PCB-99	7.5			
PCB-101	7.7	7.98	8.11	7.0 / 7.0

<sup>a</sup> from two different experiments.

the modelled C<sub>∞</sub> and the measured C<sub>air</sub> at day 9 (Table S3), during which the active air sample was taken.

PCB-8, the only dichlorinated congener analyzed, had a clear curvilinear shape and reached equilibrium within the 31 days of the experiment (Fig. 1). This was confirmed when a curve was fitted using Eq. 1, and the time-to-equilibrium (t<sub>95</sub>) was calculated. From Eq. 2 and 3, the approximate duration of the linear (kinetic) phase (t<sub>25</sub>) and estimated equilibrium concentration (C<sub>∞</sub>) were also calculated (Table 1). t<sub>25</sub> was below 7 days for PCB-8, 18, 28, and 31, but of the tri-chlorinated PCBs, only PCB-18 was close to equilibrium at the end of the experiment. The tendency for curvilinear shape decreased with increasing chlorination and for PCB-105 and higher chlorinated congeners, it became difficult to detect any curvilinear behavior within the 31 days, thus for these it was not possible to make meaningful estimates of the parameters in Table 1. For PCB-180, too many samples were below the detection limit to provide an accurate description of the uptake.

### 3.2. Field study

Wristbands from four participants were either lost or did not comply with the guidance for deployment or return. Furthermore, three persons in the exposed group did not fulfill the criteria of full-time residence in the contaminated apartment, and two persons could not fit the wristbands, which instead were worn as a pendant on the clothes; as such, all five were excluded from further data treatment. Thus, wristbands from 64 participants in the exposed group and 21 in the reference group, living in 52 and 18 different apartments, respectively, were included in the data analysis. Information on hours spent in apartment was missing from 15 of the included participants.

PCBs were detected in all wristbands, including those worn by the reference group. The congener pattern were similar to that reported in air (Andersen et al. 2020), though more higher chlorinated congeners were detected on the wristbands. Detection frequencies for the individual congeners in each group are given in Table S4. The levels in the two groups were very different, with median levels of summed measured PCBs (Σ<sub>16</sub>PCB) on the wristbands of 4086 ng/g silicone in the exposed group and 59.8 ng/g silicone for the reference group. The difference between exposure groups was significant for all congeners except PCB-153 and 180 (Fig. 2). The wristbands were dominated by the lower chlorinated congeners, which is in agreement with previous

observations for indoor air samples from the same apartments (Andersen et al. 2020), with PCB-18, 28, 31 and 52 detected at the highest levels in both the exposed and reference groups. However, low levels of PCB-118, 138, and 153 were also detected in the majority of samples (all > 70%), and PCB-180 was detected in approximately 40% of the samples from both groups.

A generic field sampling rate, R<sub>f</sub>, was also determined (Table 2) using Eq. (4), the measured air concentration in the individual apartments of the exposed group (Andersen et al. 2020), and sampling time; details on congener specific sampling rates can be found in Table S2. Most exposed participants (n = 50) recorded the actual number of hours spent in their apartment, so field sampling rate was reported both as apparent (full deployment time) as well as time-adjusted (only reported hours in apartment) field sampling rate. The mean age of exposed participants was 62.4 years (Frederiksen et al. 2020), and a large proportion was retired; thus, on average they reported staying in their apartment 79% of the time during the 7 days of sampling. The variation in field sampling rate was quite large (RSD > 50%) among the participants, but the averages for the actively worn wristbands were similar to what was observed in the kinetic study (Table 2). The apparent field sampling rate (not adjusted for time at home) was 2.6 m<sup>3</sup>/d for the exposed group. However, when adjusting for time spent at home, the overall average field sampling rate increased to 3.5 m<sup>3</sup>/d, corresponding to an MTC of 26 m/h. The average sampling rate for the static wristbands left on tables was 0.57 m<sup>3</sup>/d, corresponding to an MTC of 4.2 m/h. A summary of the different types of generic sampling rates and MTCs can be found in Table 2.

### 3.3. Exposure estimates from wristbands

The current study also had the purpose to test the potential of the wristbands to be used for ranking of PCB exposure in both domestic and occupational settings and possibly even for semi-quantitative exposure estimates, in a setting where the main PCB exposure source was indoor air. Fig. 3 shows the Σ<sub>16</sub>PCB concentrations on wristbands grouped according to the concentration of PCB<sub>total</sub> in air based on data presented in aggregated form in Andersen et al. (2020), both with and without adjustment for the reported time spent at home. The group < 300 ng/m<sup>3</sup> comprised the reference group. There was an apparent increasing trend of PCB concentrations measured on wristbands compared to air

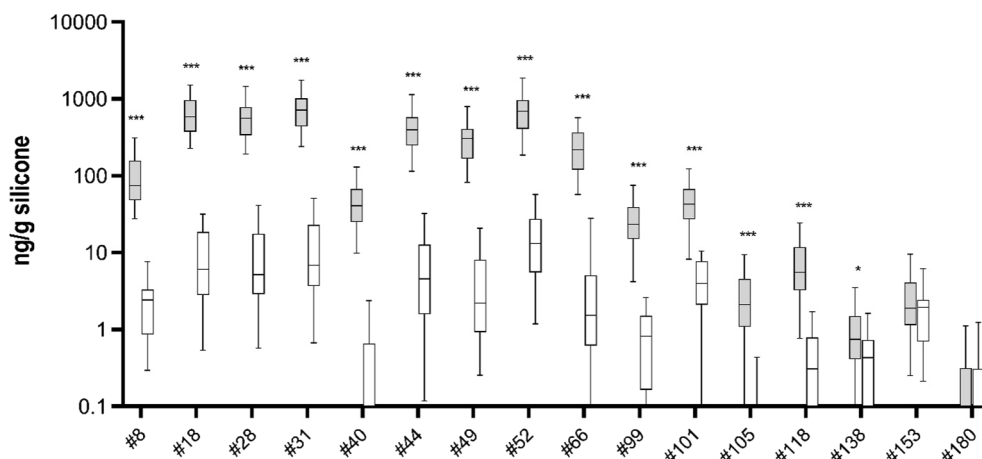


Fig. 2. Interquartile range (w. 5/95th interval) PCB levels in wristbands from the field study [ng/g silicone]. Grey: exposed group (n = 64); white: reference group (n = 21). Significance levels: \*\*\* p < 0.001; \* p < 0.01 of Mann-Whitney test. The reference group included a few non-detects for some congeners, which were set to zero.

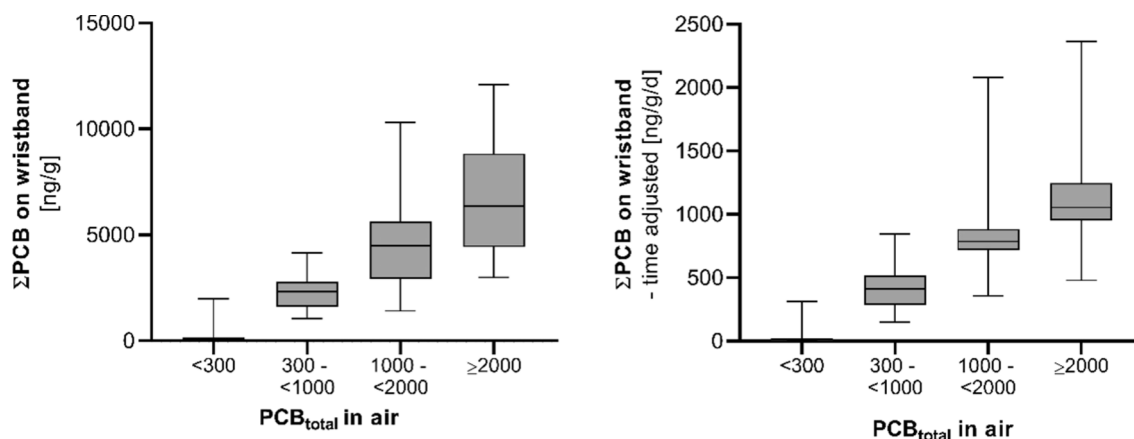


Fig. 3.  $\Sigma_{16}$ PCB on wristbands (median and 5/95th percentile) by groups of indoor air  $PCB_{total}$  level [ng/m<sup>3</sup>]. Left: Non-adjusted wristband levels (n = 85) after 7 days of sampling. Right: Wristband levels adjusted for hours spend at home and normalized to 24 h (n = 70).

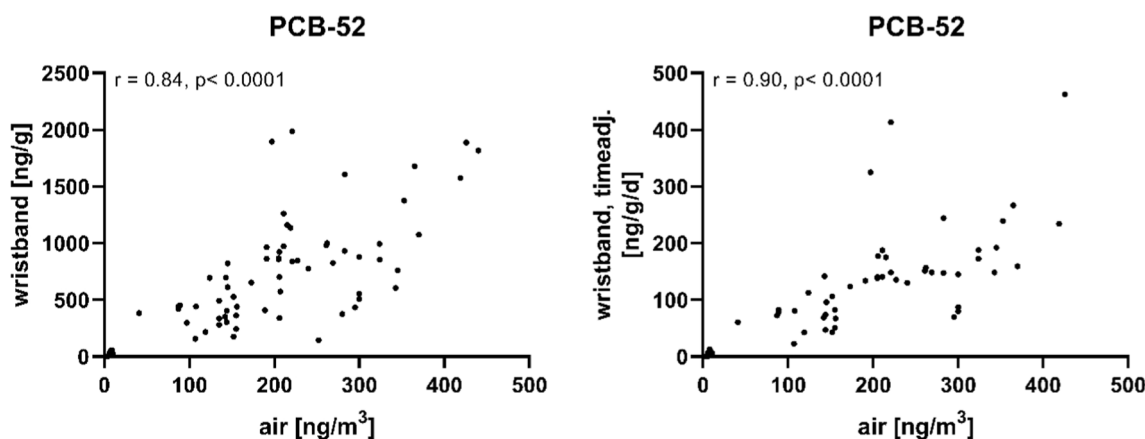


Fig. 4. PCB-52 on wristbands vs air concentration. Left: Non-adjusted wristband levels (n = 85). Right: Wristband levels adjusted for hours spend at home and normalized to 24 h (n = 70).

concentration, regardless of time-adjustment, and the groups were significantly different (Kruskal-Wallis tests, p < 0.0001). Adjusting for effective sampling time (i.e. recorded time at home) generally focused the groups; however, a few samples were adjusted away from the center of the group. The same pattern was observed when looking at the

persons and congeners individually and highly significant correlations were observed for both non-adjusted and time-adjusted wristband levels with indoor air levels for all congeners from PCB-8 to -101 (Spearman rank r < 0.0001) (Fig. 4 and S2).

## 4. Discussion

### 4.1. Air sampling kinetics and partitioning

In the current study, we determined a generic field sampling rate and mass transfer coefficient for silicone wristbands worn by human subjects. The current exposure situation was well-suited for this purpose, as the PCB exposure in the apartments was high and well-characterized; furthermore, the participants spent the majority of their time at home, and exposure from other sources was minimal. This also decreased the potential effect of bringing the wristbands to a clean environment, which could lead to some loss due to outward flux of PCBs.

Only few studies have investigated the kinetics of the sampling process of silicone wristbands (O'Connell et al., 2021; Tromp et al. 2019). The sampling rate in the current kinetics study was comparable to, though slightly lower than the previously reported generic sampling rate for SVOCs on wristbands of  $7.6 \pm 1.3 \text{ m}^3 \text{ d}^{-1} \text{ dm}^{-2}$  in a chamber with high air velocity (Tromp et al. 2019). The air velocity is important for the sampling rate as an increase in air velocity causing an increase in turbulence in the air will both increase the transport rate to the boundary layer and decrease the thickness of the boundary layer, thereby increasing the sampling rate. Vorkamp et al. (2016) showed that air velocity had a marked, non-linear effect on the sampling rates of PCBs in silicone from indoor air. In the current kinetics study, the air velocity was elevated and kept constant by a fan, but the actual value was not measured as the main purpose was to prevent the air from being stagnant. The indoor air PCB levels were expected to be very stable in the vacated apartment due to stable temperature and good agreement with previous air measurements; this is supported by the limited diurnal variation in air concentrations observed within a subgroup of inhabited homes with consecutive 4-hour samplings (Andersen et al. 2020). However, as no measures were applied to control the indoor air concentrations, in a way comparable to chamber studies, small fluctuations in the air concentration compared to the measured level at day 9 may have had a minor influence on the estimated sampling rates.

In field sampling, arm movement increases the local air velocity over the wristband. This was illustrated by the static sampling with wristbands left on a horizontal surface in the living room of five participants. The sampling rates were on average 6 times lower for the static wristbands left on a surface compared to the actively worn wristbands. Tromp et al. (2019) reported an overall average sampling rate of static passive samplers indoors from a number of different field studies of  $1.0 \pm 0.8 \text{ m}^3 \text{ d}^{-1} \text{ dm}^{-2}$ , which is in line with the sampling rate of the static wristbands in the current field study ( $0.57 \text{ m}^3 \text{ d}^{-1}$  or  $1.01 \text{ m}^3 \text{ d}^{-1} \text{ dm}^{-2}$  (Table 2)). The good agreement with previous studies supports the premise of air-side restricted uptake leading to a generic sampling rate for SVOCs (Shoeib and Harner 2002; Tromp et al. 2019). The estimated field sampling rate of worn wristbands was very close to the active air sampling rate of  $2.7 \text{ m}^3 \text{ d}^{-1}$  used by Andersen et al. (2020). The sampling rate was determined using samples from day 7, despite the fact that this exceeded  $t_{25}$  for some congeners; however, the sampling rate calculated for PCB-8 at day 7 was still among the highest among congeners (Table S2) indicating that  $t_{25}$  may be a conservative estimate of length of the linear uptake phase. The generic sampling rate was based on the congeners up to PCB-101, since air levels of the remaining congeners were very low and for higher chlorinated congeners a larger fraction will be particle bound ( $\log K_{oa} > 10$ , (Shoeib and Harner 2002)).

Based on the assumption of air-side controlled uptake, the obtained field sampling rate may apply to other gas-phase SVOCs and other wristband studies, as dimensions tend to be similar for wristbands across studies. However, the interpersonal variation was rather high and likely influenced by differences between participants (e.g. in activity level, tightness of the band around the wrist and coverage by clothes). The activity level, particularly arm movement, will have a major influence on the sampling rate by changing the local air velocity, as discussed above. The tightness of the wristband will also influence the sampling

rate, with the worst case being the wristband sitting tight to the skin and thus lowering the effective surface area of the sampler by almost 50%. Initial attempts were made to insulate the wristbands on the inside to avoid differences in effective surface area, but we did not find a good, applicable solution to this. This issue of potential sampling from the skin is not yet very well described in the literature. When the wristbands are tight to the skin, the inner layer will instead sample from the boundary layer of the skin which again is sampling from the air; thus, it is uncertain how large the effect of a reduced surface area is in practice. In the current study, the participants wore the wristbands for 7 full days, so it had to be comfortable to wear, and participants were only asked to wear it if it was not sitting tightly on their wrist. Finally, different preferences for tight/loose or long/short sleeved shirts will cause differences in the degree of coverage by clothes. Coverage may result in restricted air movement and shielding from the contaminated air; however, a study by Morrison et al. (2018) indicates that the clothes will also be contaminated with PCB and may contribute to continued exposure when in a clean environment. Many of these issues with factors potentially affecting sampling rates could be overcome by using Performance Reference Compounds (PRCs), as are commonly used in passive sampling of contaminants in water (Booij et al. 2006); however, compounds with no ethical concern in a human study would need to be used.

Partitioning coefficients were estimated for nine congeners. There was good agreement with previously published values by Gilbert et al. (2016) and Tromp et al. (2019), but these were all higher than values reported in Vorkamp et al. (2016) (Table 3), who described differences in polymer materials and discrepancy between calibration and sampling as a potential explanation for the observed difference. As is often the case in wristband studies, the silicone in the present study was not well-defined as the wristbands were designed for commercial purposes. Nevertheless, the estimated partitioning coefficients showed good correlation ( $R^2 = 0.94$ ) with the octanol-air partitioning coefficient ( $K_{oa}$ ) (Fig. S1), which may allow extrapolation to other PCBs.

### 4.2. Wristbands as personal passive samplers

Silicone wristbands have not yet been used to quantitatively estimate exposure to PCBs, though there are several studies under controlled settings on uptake rate and stability of VOCs and SVOCs in wristbands (Anderson et al. 2017; O'Connell et al., 2021; Tromp et al. 2019). However, we know from previous analyses of air and blood samples that the participants of this study were highly exposed to PCBs (Andersen et al. 2020; Frederiksen et al. 2020). Comparing the wristbands to air samples, it was clear that the wristbands worked very well as a ranking tool of exposure from air (Fig. 3) and in the current case also as a semi-quantitative tool for airborne exposure (Fig. 4 and S2). Adjusting to reported time spent at home only improved the correlations marginally, while most of the samples were more centered there were some more extreme outliers resulting in similar RSDs regardless of time adjustment. Given that the participants already spent the majority of their time in the exposed environment, the effect of some hours away may be minimal, and reporting of the hours away is probably quite crude. In addition, the contaminated clothes may also prolong the exposure outside the building, as discussed above. The wristbands have the advantage of integrating exposure over time, making them less sensitive to short-term fluctuations, and closer to actual average exposure than other more cross-sectional measures such as hand wipes or air samples. Considering the low acute toxicity of PCBs and their persistence and accumulation, time-integrating exposure might be a more important parameter to consider than short-term air measurements. In addition to reflecting PCB exposure from inhalation, the wristbands may also be a proxy for dermal exposure following absorption from air. Though the PCBs in the current study are mainly present in the gas phase, some particle deposition on the wristbands is expected, which may be relevant for less volatile SVOCs (Hammel et al. 2018).

In order to obtain reliable exposure ranking or estimates, it is



important that the wristbands either sample in the linear phase for the entire deployment period or are deployed for long enough to reach equilibrium for all compounds. However, the latter will not be feasible for PCBs as the kinetic study indicated that even with relatively high air velocities, the  $t_{95}$  was 18 days for the most volatile of the measured PCBs. The linear phase was at least 2 days for PCB-8 and 3–6 days for PCB-18, 28 and 31, which are the highly abundant tri-chlorinated congeners; all remaining congener exhibited  $t_{25}$  values greater than seven days. Although an exposure of seven days was chosen for the field study, the obtained field sampling rate was very similar to the one obtained under controlled conditions. Moreover,  $t_{25}$  is a crude estimate of the length of the linear phase, thus it may continue somewhat beyond this point. In other studies using wristbands for SVOC sampling, deployment time is often 5–7 days (Hammel et al. 2020; Hammel et al. 2018; Kile et al. 2016; Romanak et al. 2019) though some are shorter, e.g. 8–72 h (O'Connell et al., 2014; Quintana et al. 2019; Wang et al. 2019; Wang et al. 2020). Thus, if a broad range of PCBs should be assessed, a sampling time of 5–7 days seems suitable.

The advantages provided by the wristbands in form of easy deployment and handling are significant, though the limitations should also be acknowledged. Due to the nature of the wristbands sampling from air, surface contact, and skin, it can be difficult to translate the exposure measured by a wristband to an exact indoor air level. Sampling rates are often uncertain for passive samplers, however, the indoor air setting provides relatively constant temperatures, and wristbands ensure a relatively high air velocity near the sampler surface. Nevertheless, the estimated field sampling rate provided in this study will give some indication of the magnitude of exposure. A number of studies have found wristbands to be good predictors of actual personal exposure possibly because of the combined sampling pathways and extended sampling period; therefore, they have great value for ranking or even semi-quantitative purposes and not least for linking exposure to health effects or markers of effect. The good compliance from the participants also qualifies for future use.

## 5. Conclusion

Silicone wristbands were found to not only be a reliable ranking tool for PCB exposure, but also to provide a proxy for airborne exposure in PCB-contaminated indoor environments. Furthermore, the sampling kinetics of the wristbands under both controlled and field conditions were investigated, determining generic sampling rates as well as partitioning coefficients between air and silicone, which can be used for semi-quantitative estimates of external exposure based on wristband data.

Relationships to the other sampled matrices, demographics and questionnaire data are to be explored further in upcoming investigations.

### *CRedit authorship contribution statement*

**Marie Frederiksen:** Conceptualization, Methodology, Formal analysis, Investigation, Funding acquisition, Writing – original draft. **Helle Vibeke Andersen:** Conceptualization, Funding acquisition, Writing – review & editing. **Soffie Lillelund Ovesen:** Methodology, Investigation, Writing – review & editing. **Katrin Vorkamp:** Writing – review & editing. **Stephanie C. Hammel:** Writing – review & editing. **Lisbeth E. Knudsen:** Conceptualization, Writing – review & editing.

### 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 material

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