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Joint Sealants: An Overlooked Diffuse Source of Polychlorinated Biphenyls in Buildings

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In October 2000, joint sealants containing polychlorinated biphenyls (PCB) were discovered in various public buildings in Switzerland. Triggered by this event, a nationwide comprehensive study was initiated by the Swiss Agency for the Environment, Forests, and Landscape, and 1348 samples of joint sealants as well as 160 indoor air samples from concrete buildings erected between 1950 and 1980 were analyzed. Out of 1348 samples, 646 (48%) contained PCB. In 279 (21%) samples, PCB concentrations of 10 g/kg and more were detected, and concentrations of 100 g/kg of PCB or more were found in 129 (9.6%) samples. These data indicate that PCB were widely used as plasticizers in joint sealants in Switzerland. In buildings constructed between 1966 and 1971, one-third of all joint sealants investigated contained more than 10 g/kg of PCB. PCB concentrations exceeding the limit of 0.050 g/kg above which material is required to be treated as PCB bulk product waste were reached by 568 samples (42%). PCB with a chlorine content between 45 and 55%, corresponding to mixtures such as Clophen A50, Aroclor 1248, and Aroclor 1254, were encountered in 316 samples (70%). In 42 cases (26%) where joint sealants containing PCB were present, clearly elevated PCB indoor air concentrations above 1 μg/m^3 were encountered. In eight cases (5%), levels were higher than 3 μg/m^3. The Swiss tentative guideline value of 6 μg/m^3 (based on a daily exposure of 8 h) for PCB in indoor air was exceeded in one case (0.6%). On the basis of this work, representing the first large-scale nationwide analysis of the issue of PCB-contaminated joint sealants, we estimate that there are still 50−150 t of PCB present in these materials, acting as diffuse sources. They are distributed over many hundreds of buildings all over the country and represent a significant but frequently overlooked inventory of PCB. In light of the Stockholm Convention on persistent organic pollutants that entered into force last year, reduction of the release of PCB from these widely used materials is an important issue to be addressed.

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Introduction

Polychlorinated biphenyls (PCB) are toxic, resist degradation, accumulate in terrestrial and aquatic ecosystems, and are subjected to long-range transport. PCB were used in heat-transfer fluids, dielectric fluids, hydraulic fluids, paints, coatings, ceiling tiles, and sealants. PCB cause a variety of adverse health effects (*1*). PCB have been shown to cause cancer in animals, as well as several serious, noncancer, health effects on the immune system, reproductive system, nervous system, and the endocrine system. Studies on humans provide supportive evidence for the potential carcinogenic and noncarcinogenic effects of PCB.

Switzerland banned the use of PCB in "open systems" such as printing inks, sealants, lubricating and cutting oils, plastics, paints, and varnishes in 1972. The use of PCB in open systems was banned in many other industrialized countries following the recommendation of the Organization for Economic Cooperation and Development (OECD) from February 20, 1973 to its member states. In the United States, PCB production was halted in 1977. Nevertheless, more than 30 years later, materials acting as open sources of PCB are still present in surprisingly large quantities. While large efforts were made in the past 20 years to phase out "closed systems" containing PCB such as transformers and large capacitors (*2*), open systems, being more complicated to deal with, received less attention. Open systems include polysulfide-based elastic joint sealants produced between 1955 and 1975, containing significant amounts of PCB added as a plasticizer. These materials exceed the limit of 0.05 g/kg PCB, issued by the U.S. EPA and the Swiss federal authorities, above which material is required to be treated as PCB bulk product waste, by several orders of magnitude. Therefore, concrete buildings containing elastic joint sealants represent a significant inventory of PCB. In an eight-story building in Sweden, investigated by Sundahl and co-workers (*3*), the length of the joint sealants was reported to be about 4000 m, corresponding to about 1.5 t of sealants. At an average PCB content of 60 g/kg, this building represents a reservoir of 90 kg of PCB. The restoration of a five-story school building in the city of Chur, Switzerland, constructed between 1966 and 1967, revealed more than 8000 m of joint sealants, containing PCB in concentrations between 27.4 and 583 g/kg.

Despite the fact that several reports on the extent of this problem have been published (*3*−*9*), joint sealants are still not widely recognized as important diffuse sources of PCB, representing a potential hazard for human health and the environment. Most work on joint sealants containing PCB has been published by researchers in Germany, dealing mostly with the problem of indoor air contamination (*10*−*15*). Very recently, Herrick and co-workers (*16*) reported on an investigation of buildings in the Greater Boston area. Out of 24 buildings, 8 buildings contained caulking materials with a PCB content exceeding 0.05 g/kg by weight, demonstrating that the use of caulking materials containing PCB was also

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a common construction practice in the United States. The highest PCB concentration reported by Herrick and coworkers (*16*) was determined in a sample of caulking material taken from a university student housing building, containing 36.2 g/kg of PCB.

Indoor air quality might be severely affected by the presence of joint sealants containing PCB (*10*−*15*, *17*). Total indoor air PCB concentrations up to several 1000 ng/m^3 have been reported, depending on the air-exchange rate, the amount of joint sealants in a room, as well as their PCB type and content (*10*, *13*, *17*). However, even without distinct PCB sources being present, indoor air concentrations are typically more than an order of magnitude higher than corresponding outdoor levels (*18*, *19*) because of widespread diffuse secondary PCB contamination (e.g., from electrical appliances, paints, coatings, and sealants). MacLeod (*18*) reported typical indoor PCB concentrations between 0.039 and 0.580 μg/m^3 for homes, compared to outdoor levels between 0.004 and 0.018 μg/m^3. If indoor PCB sources are present and air-exchange rates are low, indoor air PCB concentrations over 10 μg/m^3 may occur (*13*, *18*). During removal of joint sealants, indoor air concentrations up to 120 μg/m^3 have been reported (*3*). Therefore, special care is required during remediation work when joint sealants containing PCB are present (*20*, *21*).

Inspired by reports about PCB-containing joint sealants in Germany, a first investigation of joint sealants in 29 public buildings has been carried out in three cantons in East and Central Switzerland between October 1999 and summer 2000. PCB concentrations between 0.022 and 216 g/kg were detected in 17 of 29 samples in this survey. Triggered by these findings, a nationwide survey initiated by the Swiss Agency for the Environment, Forests, and Landscape was started, combined with an interlaboratory study on the determination of PCB in joint sealants. The goals of this work were a nationwide, comprehensive assessment of the abundance of joint sealants containing PCB present in Swiss buildings as well as an assessment of the impact of these materials on human health and the environment. Sampling focused on concrete buildings erected between 1950 and 1980, and 1348 samples of joint sealants as well as 160 indoor air samples were analyzed.

To our knowledge, this is the first large-scale, nationwide study addressing the problem of joint sealants containing PCB present in buildings as an issue concerning both health and the environment. In the framework of these activities, a tentative guideline value for the maximum concentration of PCB in indoor air (*17*, *22*, *23*) was established (6 μg/m^3 based on 8-h exposure, e.g., public buildings and 2 μg/m^3 based on 24-h exposure, e.g., homes) and guidelines assisting building owners and contractors on how to identify and, if necessary, safely remove joint sealants containing PCB (*20*, *21*) were published.

Experimental Section

Sampling Strategy. In cooperation with local authorities and building owners, 1348 samples of joint sealants and 160 indoor air samples from Swiss concrete buildings erected between 1950 and 1980 were collected and analyzed by selected laboratories. Sampling techniques, analytical methods, and requirements for documentation were discussed and harmonized, and an interlaboratory study on the determination of PCB in joint sealants was started in cooperation with the participating laboratories at an early state of the project (*20*, *22*).

PCB Analysis. Joint Sealants. Each participating laboratory which fulfilled the requirements of the interlaboratory study (see section Quality Assurance) was qualified to use its own analytical methods based on gas chromatography/mass spectrometry (GC/MS) or gas chromatography/electron capture detection (GC/ECD). A PCB detection limit (d.l.) of 0.02 g/kg for the total PCB content was requested. For most of the samples, the detection limit for individual PCB congeners was 0.002 g/kg or lower. To improve comparability, results were reported as concentrations of the individual PCB congeners PCB-28, -52, -101, -138, -153, and -180. The total PCB content was estimated as the sum of these congeners multiplied by a standard factor of 5 (*24*) and the type of the PCB mixture was determined by comparison to PCB reference materials. Wavelength-dispersive X-ray fluorescence spectrometry (WD-XRF) was investigated as a rapid screening method for joint sealants suspected to contain PCB. Samples were analyzed without further preparation on a WD-XRF spectrometer (PW 2400, Philips). Detection limits were between 0.01 and 0.1 g/kg, depending on the actual element. On the basis of long-term experience with routine analysis of chlorine in mineral oil samples, the detection limit for chlorine in mineral oils, a roughly comparable matrix, was estimated to be 2−5 ppm (0.002−0.005 g/kg). This translates into a detection limit for PCB of 0.01−0.025 g/kg (on the basis of a PCB with a chlorine content of 20%). The ubiquitous presence of chlorine as an additive or contaminant in many building materials, however, may lead to significantly higher detection limits in some cases. Selected samples were analyzed for chlorine and lead with independent methods (ion chromatography and graphite furnace atomic absorption spectrometry) to verify the WD-XRF results.

Indoor Air. Determination of PCB in indoor air was based on low-volume air sampling and analysis by GC/MS, as reported previously (*17*). The total PCB content for an indoor air sample was estimated as the sum of the six PCB congeners PCB-28, -52, -101, -138, -153, and -180 multiplied by a factor of 5 (*24*).

Quality Assurance. PCB Analysis in Joint Sealants. For the interlaboratory study, two samples of joint sealants known to contain PCB were randomized and sent to each of the participating 17 laboratories. After all results were received, the adjusted *z*-scores were calculated and two laboratories were identified as outliers. On the basis of the results of the remaining laboratories, the corrected mean total PCB concentrations of the two round robin samples were 117 and 141 g/kg. Coefficients of variance were ±2.9% and ±5.4%. A comparison of the analytical methods of the individual laboratories and the results submitted revealed no significant differences between extraction methods (Soxhlet, ultrasound, accelerated solvent extraction), solvents (acetone, hexane, cyclohexane, *n*-octane, or toluene), cleanup methods (no cleanup versus filtration using Na_2SO_4), and detection methods (GC/MS versus GC/ECD). Digesting the polymer matrix with oleum prior to extraction led to a reduction of the recoveries by up to 30% ($n = 60$ determinations) because of partial dechlorination of PCB-congeners PCB-28 and -52 and, to some extent, higher chlorinated PCB-congeners.

PCB in Indoor Air. Analytical methods were validated by parallel measurements of indoor air concentrations among the participating laboratories. Coefficients of variance between 5 and 25% were determined. On the basis of a sampling volume of 180 l, typical blank levels for the individual PCB congeners PCB-28, -52, -101, -138, -153, and -180 were between 0.1 and 1 ng/m^3 (*17*). To ensure coherent measuring procedures between all laboratories involved, instructions on how determinations of PCB in indoor air should be carried out were issued (*25*). These are briefly summarized as follows: All laboratories were instructed to air the rooms the evening before sampling took place and to keep windows and doors shut overnight and during air sampling. Measurements were to be carried out at conditions as close as possible to the normal use of the room to assess the effective exposure of people present in the respective rooms. The hourly flow of the air-sampling equipment was to be adjusted to be below

TABLE 1. Temporal Distribution of PCB Concentrations (c = [g/kg]) in 1348 Joint Sealant Samples from Public Buildings in Switzerland

	number of samples [n]	number of samples [n] c < d.l.[a]	number of samples [n] d.l. ≥ c < 0.1 g/kg	number of samples [n] 0.1 ≥ c < 10 g/kg	number of samples [n] c ≥ 10 g/kg	percentage of samples [%] c ≥ 10 g/kg
before 1950	1	1	0	0	0	0
1950−54	6	5	1	0	0	0
1955−59	46	38	0	5	3	7
1960−61	27	17	1	6	3	11
1962−63	50	23	10	5	12	24
1964−65	52	25	9	5	13	25
1966−67	91	40	2	15	34	37
1968−69	114	44	9	20	41	36
1970−71	176	64	19	37	56	32
1972−73	257	152	22	49	34	13
1974−75	195	135	20	36	4	2
1976−77	28	21	2	2	3	11
1978 and later	33	18	10	5	0	0
unknown age	272	119	23	54	76	28
total	1348	702	128	239	279	21

[a] d.l.: detection limit; for most of the samples, the limit of detection for individual PCB congeners was 0.002 g/kg or lower and 0.02 g/kg for total PCB content.

10% of the room volume. Priority was given to rooms with low air-exchange rates where people stay for extended periods.

Results and Discussion

PCB in Joint Sealants: Occurrence and Inventory in Switzerland. Occurrence of Joint Sealants Containing PCB in Public Buildings in Switzerland. Sampling focused on concrete buildings erected between 1950 (estimated start of use of joint sealants containing PCB as a plasticizer) and 1980 (eight years after the use of PCB in open applications has been banned in Switzerland). From buildings all over Switzerland, samples of joint sealants were collected and analyzed. Out of 1348 samples, 646 (48%) contained PCB. In 279 samples (21%), PCB concentrations exceeded 10 g/kg, indicating that PCB were widely used as plasticizers in joint sealants in Switzerland.

Samples were classified into yearly intervals between 1960 and 1977 (5-year intervals between 1950 and 1959) and sorted into three concentration ranges (Table 1). The graphical representation of these data is shown in Figure 1. The first series (black bars) represents the number of samples of joint sealants containing at least 10 g/kg of PCB, peaking between 1970 and 1971. The number of samples containing less than 10 g/kg of PCB (gray and white bars) peaks slightly later in the time period between 1972 and 1973. Most likely, PCB were not added as plasticizers to joint sealants with PCB concentrations below 10 g/kg, since PCB need to be added in the percent range to maintain the elasticity of the material. These samples are likely to reflect PCB contamination, for example, because of the use of the same equipment for formulation or application of caulking materials with PCB and with alternative non-PCB plasticizers (e.g., chlorinated paraffins) or because of secondary contamination by migration of PCB (originating from joint sealants containing PCB replaced previously) from adjacent construction materials.

Probability to Encounter Joint Sealants Containing PCB on the Basis of the Year of Construction. On the basis of the total number of samples analyzed in each 5-year interval, the percentage of samples containing more than 10 g/kg of PCB was calculated for each interval, representing the probability to encounter these materials in a concrete building from the respective period (see last column of Table 1). In buildings constructed between 1966 and 1971, one-third of all joint sealants investigated contained more than 10 g/kg of PCB. In buildings constructed before 1955 and after 1978, however, no joint sealants containing more than 10 g/kg of PCB were encountered.

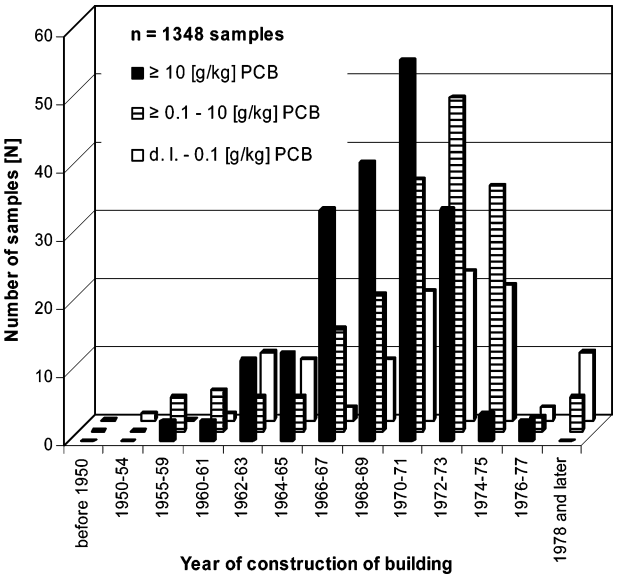

FIGURE 1. Number of joint sealants analyzed in three concentration ranges (d.l. = detection limit, 0.02 g/kg PCB).

Concentration Range of PCB in Joint Sealants in Public Buildings in Switzerland. The PCB concentrations determined in all 1348 samples were sorted in six concentration ranges, spanning the range between the detection limit and concentrations above 100 g/kg of PCB, as shown in Table 2. Most samples containing PCB were encountered in the concentration ranges above 0.1 g/kg of PCB. There was no preference for a particular concentration range; about 10% of all samples were found in each of the four ranges above 0.1 g/kg of PCB. PCB concentrations exceeding the limit of 0.050 g/kg above which material is required to be treated as PCB bulk product waste were reached by 568 (42%) of the samples. In 279 (21%) samples, high PCB concentrations of 10 g/kg and more were detected, and more than 100 g/kg of PCB were found in 129 (9.6%) samples. The maximum PCB concentration encountered was 550 g/kg, that is, more than half of the mass of that particular joint sealant represented PCB, added as a plasticizer.

Type of PCB Mixtures in Joint Sealants in Public Buildings in Switzerland. The type of PCB mixture present

TABLE 2. Distribution of PCB Concentrations in 1348 Joint Sealant Samples from Public Buildings in Switzerland

PCB content [g/kg]	number of samples [*n*]	percentage [%]
<d.l.[a]	702	52
>d.l.	646	48
d.l. to <0.05	78	5.8
0.05 to <0.1	50	3.7
0.1 to <1	122	9.1
1 to <10	117	8.7
10 to <100	150	11.1
>100	129	9.6
total	1348	100

[a] d.l.: detection limit; for most of the samples, the limit of detection for individual PCB congeners was 0.002 g/kg or lower and 0.02 g/kg for total PCB content.

in joint sealants was determined for 453 samples (193 samples could not be attributed unambiguously), as shown in Table 3. Most samples (70%) contained medium-chlorinated PCB mixtures similar to Clophen A50, Aroclor 1248, or Aroclor 1254. PCB mixtures with high chlorine content, referring to Clophen A60, Aroclor 1260, and Aroclor 1262, amounted to 20%, and only 10% of the samples contained low-chlorinated PCB mixtures, such as Clophen A30, Clophen A40, or Aroclor 1242. These data are similar to findings by Herrick and co-workers (*16*) for caulking materials taken from buildings in the Greater Boston area. In this smaller sample set, Aroclor 1254 was present in seven out of eight samples, and one sample was attributed to Aroclor 1260. Analyzing the PCB congener pattern of the samples from our study revealed that the relative concentrations of PCB-28 and PCB-52 were often lower than the concentrations of these congeners in the corresponding technical PCB mixtures, indicating partial depletion of the more volatile lower chlorinated PCB congeners in joint sealants over the years by volatilization.

To our knowledge, there are no reports in the current literature on nationwide large-scale studies on the problem of joint sealants containing PCB, as reported in this work. Nevertheless, data from other reports do agree well with our findings. In a German office building erected in 1969, Benthe and co-workers found PCB concentrations between 10 and 400 g/kg in joint sealants on the basis of Thiokol rubber (*15*). In Sweden, Sundahl and co-workers report PCB concentrations between 47 and 81 g/kg in elastic sealants (*3*), while Herrick and co-workers (*16*) detected PCB levels up to 36.2 g/kg in sealants from buildings from the Greater Boston area.

Environmental Impact. The widespread presence of PCB in joint sealants, as demonstrated by this study, raises concerns about the environmental impact of this reservoir of persistent organic pollutants (POP). Joint sealants represent a significant stockpile of PCB. According to our results, the use of these materials was a common construction practice between 1955 and 1975, and PCB are still found in many concrete buildings in Switzerland today. On the basis of figures about the recovered mass of PCB in caulking materials removed when large concrete buildings were restored (*3*), an estimated total number of large concrete buildings in Switzerland, and the data from our study, we estimate that 50−150 t of PCB are still present in joint sealants in buildings in Switzerland. For comparison, a Swedish study (*26*) estimates that 150−600 t PCB are present in buildings. The Norwegian Pollution Control Authority (SFT) estimated that 100 t of PCB were applied in joint sealants between 1963 and 1972 in Norway; only half of this amount had been disposed of until 2000 (*27*). The German Federal Environmental Agency estimated that about 20 000 t of Thiokol joint sealants with at least 2000 t of PCB have been processed in Germany (*28*). For Denmark, Wilkins and co-workers cite the Organization of Sealant Branch's Manufacturers and Distributors, reporting an estimated inventory of 75 t PCB in caulking materials (*29*). Besides volatilization into air, joint sealants containing PCB must be considered as a relevant source of emissions of PCB into soil and water as well. According to our study, PCB were present in 51% of all outdoor joint sealants (compared to 43% of all indoor joint sealants). Hellman and co-workers (*5*) showed that soil samples collected in the vicinity of an eight-story apartment building (with joint sealants containing PCB) in Finland exhibited elevated PCB concentrations in the upper soil layers (0−0.1 m and 0.1−0.3 m). The highest concentration of PCB (6.62 mg/kg soil) was determined in 0.1-m distance from the building shell in a depth of 0−0.1 m. Decreasing concentrations of PCB were found in 0−0.1 m depth at distances of 0.5 m (1.3 mg/kg soil), 1 m (0.71 mg/kg soil), 2 m (0.15 mg/kg soil), and 10 m (0.04 mg/kg soil). The authors concluded that PCB in joint sealants of the apartment building were transferred into the soil, resulting in soil concentrations above the regulatory limit (0.5 mg/kg). Therefore, it must be considered that joint sealants containing PCB in building shells may contaminate the surrounding soil by particulate losses of aging sealants and washout with precipitation.

Fast Screening for PCB in Joint Sealants on the Basis of WD-XRF. Addressing the problem of PCB-contaminated buildings on a nationwide scale required a large amount of samples to be analyzed. Wavelength-dispersive X-ray fluorescence spectrometry (WD-XRF) was investigated as a rapid screening method for joint sealants suspected to contain PCB, and a subset of 85 samples of this study was analyzed with this method. Chlorine concentrations between <1 g/kg and 200 g/kg were found, corresponding to a maximum PCB content of about 370 g/kg, calculated as Clophen A50. Next to chlorine, up to 20 g/kg lead and traces of barium, titanium, and manganese were detected in several samples. Since WD-XRF determines total chlorine concentrations, chlorine sources other than PCB, such as chlorinated paraffins, other chlorinated organic compounds, and inorganic chlorine, need to be considered. As a matter of fact, chlorinated paraffins were detected in about one-third of the samples of the subset analyzed in this study. The ubiquitous presence of chlorine as an additive or contaminant in many materials, however, may severely impair the specificity of the WD-XRF approach for samples in the low concentration range. Therefore, positive samples from WD-XRF screening need to be confirmed by GC/MS or GC/ECD. Consequently, WD-XRF is only useful as a rapid screening method if a low percentage of samples containing chlorine is anticipated.

PCB in Indoor Air—Results of Investigations in Switzerland. Joint Sealants Containing PCB and Indoor Air Quality. As a consequence of the widespread presence of joint sealants containing PCB, it was important to investigate

TABLE 3. Attribution of PCB-Containing Joint Sealant Samples ($n = 646$) to Types of PCB Mixtures

type of PCB mixture	number of samples	percentage [%]	chlorine content [%]	technical mixtures
low chlorination	46	10	30−45	Clophen A30/A40, Aroclor 1242
medium chlorination	316	70	45−55	Clophen A50, Aroclor 1248/1254
high chlorination	91	20	55−65	Clophen A60, Aroclor 1260/1262
not determined	193			

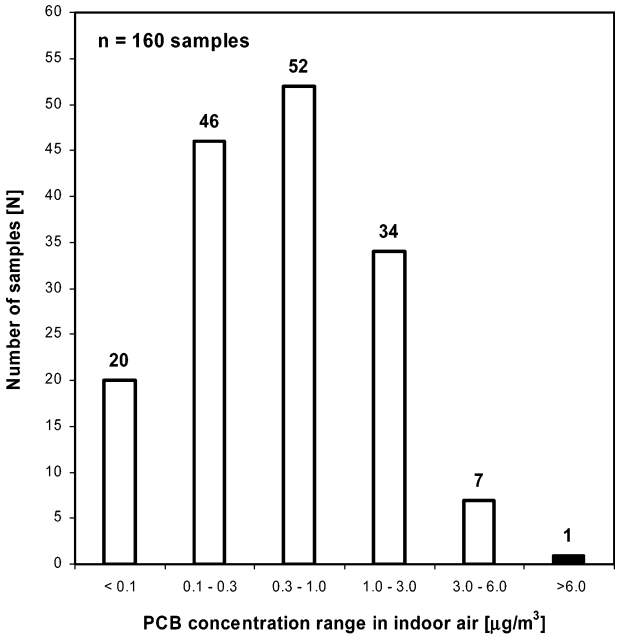

FIGURE 2. PCB indoor air concentrations determined in 160 cases where joint sealants containing PCB were present. In one case (0.6%), the Swiss tentative guideline value (based on a daily exposure of 8 h) of 6 μg/m^3 for PCB in indoor air was exceeded.

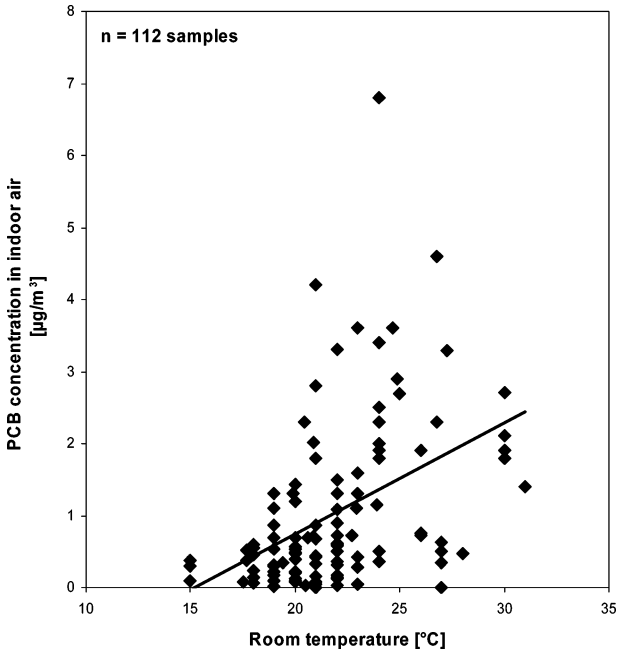

FIGURE 3. PCB indoor air concentrations determined in 112 cases as a function of the room temperature.

how the indoor air quality is affected by PCB evaporating from these materials. Figure 2 shows a histogram representing the concentrations of PCB in indoor air determined in 160 cases where joint sealants containing PCB were present. Compared to typical indoor air PCB concentrations between 0.039 and 0.580 μg/m^3, as reported by MacLeod (*18*), significantly higher concentrations were encountered in buildings where joint sealants containing PCB were present. In 42 cases (26%), elevated PCB indoor air concentrations above 1 μg/m^3 were encountered. In eight cases (5%), levels were higher than 3 μg/m^3. In one case (0.6%), the Swiss tentative guideline value of 6 μg/m^3 (based on a daily exposure of 8 h) for PCB in indoor air (*17*, *22*) was exceeded. The mean PCB indoor air concentration for all 160 sites was 0.79 μg/m^3 and the median concentration was 0.41 μg/m^3. These data show that the presence of sealants containing PCB does lead to elevated indoor air PCB concentrations. However, the Swiss tentative guideline value for the maximum tolerable PCB indoor air concentration of 6 μg/m^3 was exceeded only in one case, although joint sealants containing PCB were present in all buildings investigated.

The range of PCB indoor concentrations reported herein corresponds well with the data from other studies. Fromme and co-workers (*12*) determined indoor PCB concentrations in rooms with possible presence of joint sealants containing PCB in public utility buildings in Berlin. An average PCB indoor air concentration of 0.114 μg/m^3 (maximum 7.360 μg/m^3) was determined in community rooms of schools and child-care centers. In 5% of the schools, PCB indoor air concentrations of more than 3 μg/m^3 were encountered, the same percentage that we observed in our study. Gabrio (*13*) and co-workers detected PCB indoor air concentrations between 0.077 and 10.625 μg/m^3 in three schools where joint sealants containing PCB were present.

From our data and that which has been reported in the literature so far, we conclude that the range of PCB concentrations encountered in buildings where joint sealants containing PCB are present may easily span up to 2 orders of magnitude. As demonstrated in Figure 2, the presence of joint sealants containing PCB does not usually lead to PCB indoor air concentrations close to or above the tolerance limit of 6 μg/m^3 (based on a daily exposure of 8 h). The impact of the presence of these materials on indoor air quality is, among other factors, dependent on the air-exchange rate of the room. Typical air-exchange rates in buildings (*30*) may vary between 0.02 h^{-1} and 3.6 h^{-1}. Other parameters driving the resulting indoor air PCB concentrations include the type of PCB mixture present in the joint sealants, the surface area of joint sealants present in the respective room which is in contact with the indoor air, the temperature of the construction material adjacent to the joint sealants, and the room temperature.

On the basis of the data from our study, the PCB indoor air concentrations are shown as a function of the room temperature in Figure 3 for 112 cases. Even though this data set consists of pooled data from different buildings (instead of a repetitive data set based on individual rooms measured at different temperatures), a slight trend showing increasing PCB indoor air concentrations with increasing room temperature was observed. A positive correlation of these two variables has been reported previously by Balfanz and co-workers (*10*), on the basis of a comparison of PCB indoor air concentrations of 10 individual rooms measured in summer and winter. Therefore, the PCB indoor air concentration is also a function of the current room temperature. However, the high variability of the data indicates that next to the room temperature, other factors such as the air-exchange rate in the room and the exposure of joint sealants to direct sunlight radiation need to be considered.

Joint Sealants as Long-Term Diffuse Emitters of PCB. It is important to recognize that joint sealants represent long-term diffuse sources for PCB. Elevated concentrations of PCB in indoor air are not a transient phenomenon. The PCB inventory present in these materials is large enough to sustain elevated levels of PCB in indoor air for a very long period of time. Sundahl and co-workers (*3*) calculated, on the basis of building size and air-exchange rate, that annual emissions of 60 g PCB in an eight-story building are sufficient to sustain a permanent indoor air concentration of 0.6 μg/m^3. The total PCB inventory of 90 kg present in this building is, theoretically, sufficient to maintain elevated indoor air concentrations for many hundreds of years.

Environmental, Remediation, and Health Aspects. Within the last two decades, significant progress has been made in the phasing-out of PCB in closed applications, for example, by replacing transformers and capacitors containing PCB. In Switzerland, the inventory representing PCB in transformers could be lowered from about 1800 to 2700 t in the years between 1975 and 1985 to less than 10 t today. Nevertheless, according to the results of this study, joint sealants containing PCB are still present in many concrete buildings erected between 1955 and 1975. In buildings constructed between 1966 and 1971, one-third of all joint sealants investigated contained more than 10 g/kg of PCB. Although the use of PCB in open applications was banned in Switzerland in 1972, we estimate that there are still 50−150 t of PCB present in joint sealants, acting as diffuse PCB sources distributed over many hundreds of buildings all over the country. Joint sealants represent a major inventory of PCB. Removal and appropriate disposal of old joint sealants from construction materials is crucial to prevent significant amounts of PCB being released into the environment and, eventually, into the food chain.

The Stockholm Convention, ratified by Switzerland in 2003, requires governments to take steps to reduce the release of persistent organic pollutants (including PCB) with the goal of their continuing minimization and, where feasible, ultimate elimination. Thus, from an environmental as well as from a legal point of view, joint sealants, representing a major PCB inventory today, need to be localized, removed carefully by using suitable techniques, and disposed of as hazardous waste in high-temperature incineration plants, whenever buildings are renovated or torn down. Two guidelines (*20*, *21*) addressing these issues, including practical aspects on how to remove these materials safely, have been published recently as a result of this work. It is required that joint sealants present in buildings erected between 1955 and 1975 are to be analyzed before these structures are renovated or torn down.

Indoor air quality might be severely affected by the presence of joint sealants containing PCB. If a significant amount of these materials are present indoors, and if their PCB concentration exceeds 10 g/kg, rooms where people are present for extended periods should be checked for increased indoor air PCB levels. If the tentative guideline value (*22*) of 6 μg/m^3 for PCB in indoor air (on the basis of a daily exposure of 8 h) is exceeded, PCB sources need to be identified, removed, and disposed of as hazardous waste as soon as their PCB content exceeds 0.05 g/kg. In all other cases, the presence of joint sealants containing PCB should be documented so that they may be removed and disposed of when the building is renovated or torn down.

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