PCBs in Open Applications: Part A: Sealants/caulks in construction

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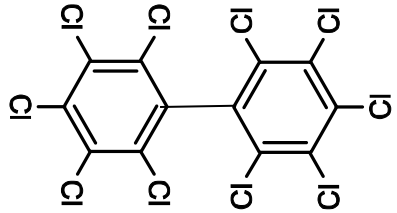
https://scholar.google.com/citations?user=-Cexto4AAAAJ&hl=en

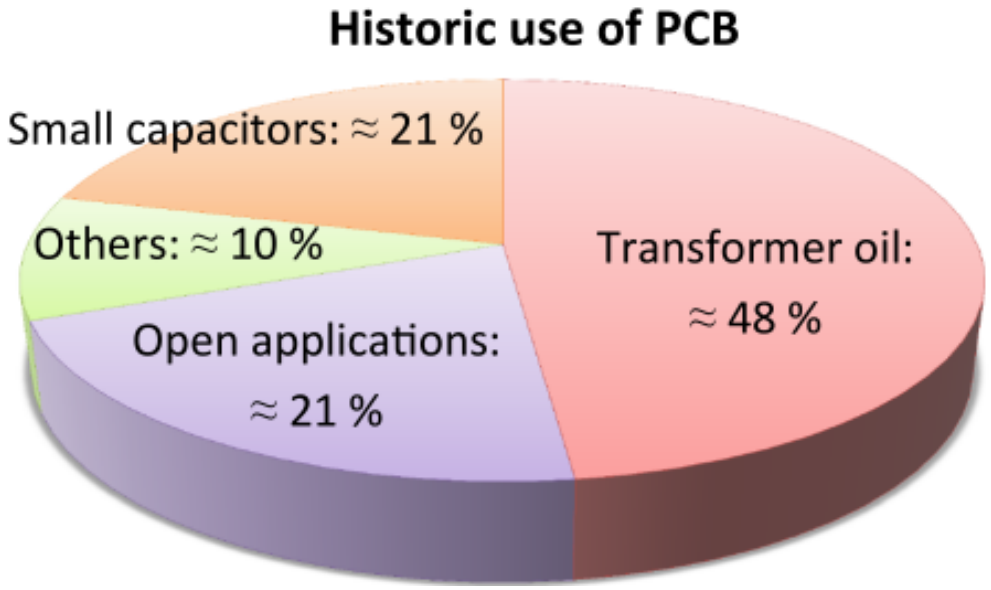
Content of Presentation

- Total use of PCB in “open” and “closed” application
- Trade names of PCBs and major producing countries
- Overview on PCB use in sealants
- Inventory experiences of selected country for PCBs and other POPs in sealants
- PCB releases from buildings and environmental contamination
- Some conclusions.

Global PCB use – “Closed“ and “open“ applications

- From the of global PCB production volume (1.3 mio t) approx. 21 % was used for open applications (ca. 300,000 tonnes).
- The most significant use of PCBs in “open applications” was in permanently elastic sealants (caulking) in buildings constructed from the 1950s to 1980.
- The use distribution of PCB in open applications depend on the region or the individual country. E.g. West Germany sealants were 80% of open PCB applications while in East Germany this use was less than 10%. The use depends e.g. on construction activities between 1950s to 1970s and the use of sealants.





Breivik, K., et al.. 2007 Science of the Total Environment,. 377, 296-307.

Trade names of PCBs

- A wide range of trade names for PCBs and for PCBs in products.
- The purchase of a country depends on the trading partners/countries.
- In Germany: West Germany mainly purchase from own producer Bayer. While East Germany purchased PCBs from Czechoslovakia 1960/70.

Abestol (t, c)	Dl(a)conal	Phenoclar DP6 (Germany)
Abuntol (USA)	DP 3, 4, 5, 6.5	Phenoclor (t, c) (France)
Aceclor (t) (France, Belgium)	Ducanol	Phenoclor DP6 (France)
Acooclor (Belgium)	Duconal (Great Britain)	Phyralene (France)
Adkarel	Duconol (c)	Physalen
ALC	Dykanol (t, c) (USA)	Plastivar (Great Britain)
Apirolia (t, c)	Dyknol (USA)	Polychlorinated biphenyl
Apirolio (t, c)	E(d)ucaral (USA)	Polychlorobiphenyl
Areclor (t)	EEC-18	Pryoclar (Great Britain)
Arochlor (t, c)	EEC-IS (USA)	Pydraul (USA)
Aroclor (t, c) (USA)	Elaol (Germany)	Pydraul 1 (USA)
Aroclor 1016 (t, c)	Electrophenyl (France)	Pydraul 11Y (USA)
Aroclor 1221 (t, c)	Electrophenyl T-60	Pyralene (t, c) (France)
Aroclor 1232 (t, c)	Elemex (t, c) (USA)	Pyralene 1460, 1500, 1501 (F)
Aroclor 1242 (t, c)	Elexem (USA)	Pyralene 3010, 3011 (France)
Aroclor 1254 (t, c)	Eucarel (USA)	Pyralene T1, T2, T3 (France)
Aroclor 1260 (t, c)	Fenchlor 42, 54, 70 (t, c) (Italy)	Pyramol (USA)
Aroclor 1262 (t, c)	Hexol (Russian federation)	Pyranol (t, c) (USA)
Aroclor 1268 (t, c)	Hivar (c)	Pyrochlor
Arubren	Hydol (t, c)	Pyroclar (Great Britain)
Asbestol (t, c)	Hydrol	Pyroclor (t) (USA)
ASK	Hyvol	Pyromal (USA)
Askarel (t, c) (USA)	Hywol (Italy/USA)	Pyronal (Great Britain)
Auxol (USA)	Inclar (Italy)	Pysanol
Bakola	Inclor (Italy)	Saf(e)-T-Kuhl (t, c) (USA)
Bakola 131 (t, c)	Inerteen 300, 400, 600 (t, c)	Safe T America
Bakolo (6) (USA)	Kanechlor (KC) (t, c) (Japan)	Saft-Kuhl
Biclor (c)	Kanechor	Sanlogol

Trade names of PCBs

- Therefore as a first step the former production of PCBs and former use of PCBs in products should be explored.
- Czechoslovakia and Russia were major producers and exporters from “East“.
- France, Germany, Italy, Japan, UK and US were major potential exporters to Serbia/Yugoslavia from “West“.

Chlorextol (t)	Kaneclor (t,c)	Sant(h)osafe (Japan)
Chlorinated Diphenyl	Kaneclor 400	Sant(h)othera (Japan)
Chlorinol (USA)	Kaneclor 500	Sant(h)othern FR (Japan)
Chlorintol (USA)	Keneclor	Santosol
Chlorobiphenyl	Kennechlor	Santoterm
Chloroecxtol (USA)	Leromoli	Santotherm (Nippon)
Chorextol	Leromoll	Santotherm FR
Clophen (t, c) (Germany)	Leronoll	Santovac
Clophen Apirorlio	Magvar	Santovac 1
Clophen-A30	Man(e)c(h)lor (KC) 200,600	Santovac 2
Clophen-A50	Manechlor (Nippon)	Santovec (USA)
Clophen-A60	MCS 1489	Santowax
Cloresil	Montar (USA)	Santvacki (USA)
Clorinol	Nepolin (USA)	Saut(h)otherm (Japan)
Clorphen (t)	Niren	Siclonyl (c)
DBBT	No-Famol	Solvol (t, c) (Russian Federation
Delorene	NoFlamol	Sorol (Russian Federation)
Delor (Czech Republic)	No-Flamol (t, c) (USA)	Sovol (Russian Federation)
DI 3,4,5,6,5	No-flanol (t,c) (USA)	Sovtol (Russian Federation)
Diachlor (t,c)	Nonflammable liquid	Terpenylchlore (France)
Diaclor (t, c)	Non-flammable liquid	Therainol FR (HT) (USA)
Diaconal	Orophen (Former East Germany)	Therminol (USA)
Dialor (c)	PCB	Therminol FR
Diconal	Pheneclor	Therpanylchlore (France)
Disconon (c)	Phenochlor	Ugilec 141, 121, 21

PCB use in sealants/caulk

- Sealants or caulk are flexible materials used to seal gaps mainly in constructions.
- PCBs were added to joint sealant materials in order to improve the flexibility of the material, increase resistance to mechanical erosion and flammability, and improve adherence to other building materials.
- Major PCB use were **polysulfide sealants**. Main brand: **Thiokol** Other commercial products: **Thioflex, Vulkseal, Vulkfil, Lasto-meric, 1K, Terostat, PRC and Rubberseal.**
- Other sealants were polyurethane, acrylic, epoxy, and butyl sealants might have been produced with PCBs (PCNs; chlorinated paraffins).
- PCBs have not been used in silicone rubber.
- PCBs in sealants have been widely used (e.g. Canada, Germany, Denmark, Finland, Norway, Sweden, Switzerland, UK, USA,).
- The use of PCBs in sealants in low- and middle-income countries has not been assessed.
- The time frame when PCB in sealants were used differ somewhat between the countries.

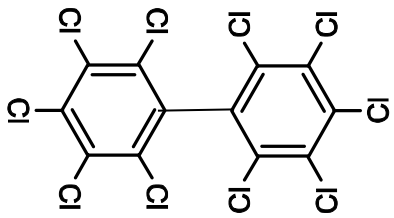
PCB concentrations in sealants/caulks

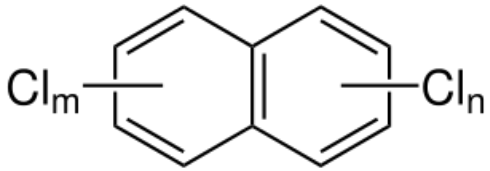
- For the production of sealants, the PCB were mixed with the polysulfide polymer at the construction site.
- PCBs concentrations in sealants are normally between 3 to 30%.
- Sealants with PCB concentration below 0.5% are rather from secondary contamination from former sealant or other primary contamination source.

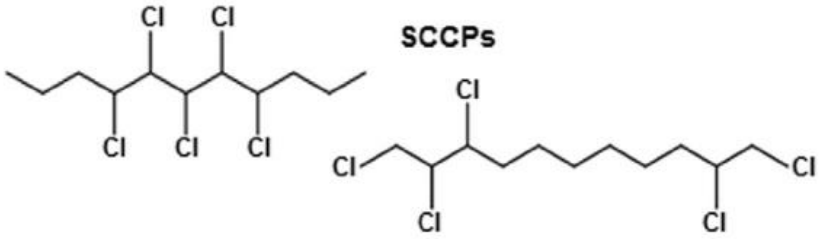
Location	Material and age	Concentration of PCBs	Source of data	Reference
Toronto, Canada	Buildings constructed from 1945 to 1980	570–82,090 mg/kg	Measured in currently in-use sealants	This study
Southern Sweden	Caulking manufactured from 1965 to 1973	Sealant is 200,000 mg/kg	Not measured, concentrations based on landfill records	Persson et al. (2005)
Boston, USA	Buildings constructed or renovated in 1970s	0.56–32,600 mg/kg	Measured in currently in-use sealants	Herrick et al. (2004)
Switzerland	Buildings constructed from 1950 to 1978	20 mg/kg–550,000 mg/kg	Measured in currently in-use sealants	Kohler et al. (2005)
Finland	Sealants manufactured from 1960 to 1975	50,000 mg/kg to 300,000 mg/kg	Not measured, concentrations based on industry information	Priha et al. (2005)
Stockholm, Sweden	Sealants, age unknown	80,000 mg/kg to 160,000 mg/kg	Not measured, concentrations based on industry information	Astebro et al. (2000)
Sweden	Building sealant, 1969	47,000 mg/kg to 81,000 mg/kg	Measured in sealants removed from building	Sundahl et al. (1999)

Other POPs additives use in sealants

- Other POPs have been used as additives in sealants including polychlorinated naphtalene (PCNs) which have been listed in the Stockholm Convention in 2015.
- Chlorinated paraffins have substituted PCBs and PCNs in 1970s. They were listed 2017 in the Convention.
- Major use of these alternatives are also polysulfide sealants but also polyurethane, acrylic and butyl sealants but not in silicone sealants.
- Minor data to which extent SCCPs, MCCPs and LCCPs were and are used as additive in what type of sealants.
- Screening technologies (XRF or “Beilstein test” copper wire & flame (fumes!)) can be used to detect chlorine as indicator. But cannot be used to distinguish between these chlorinated additives. To distinguish between the different additives instrumental analysis (GC) needed.







PCB use in sealants – screening of buildings

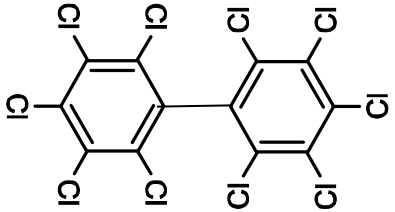
- Some countries have used PCB sealants quite extensively.
- Major use were concrete buildings (often pre-fabricated).
- **In Switzerland:** 30% of the sealants tested and 50% of the tested houses constructed 1950 to 1975 contained PCB (Kohler et al. 2005).
- **In US:** half of the school and other buildings with caulks constructed over this period are estimated to contain PCBs (Herrick et al. 2004).
- Larger concrete buildings can contain PCB in 100 kg to tonne scale in sealant materials.

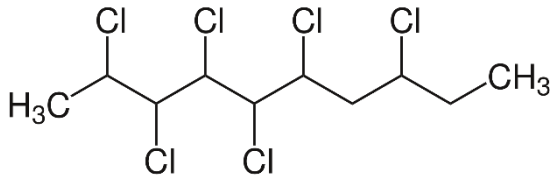
Kohler et al. (2005) Joint sealants: An overlooked diffuse source of polychlorinated biphenyls in buildings. Environ Sci Technol, 39(7), 1967-1973.

Herrick, R. F., McClean, M. D., Meeker, J. D., Baxter, L. K., & Weymouth, G. A. (2004) An unrecognized source of PCB contamination in schools and other buildings, Environ Health Perspect 112, 1051-1053.

Monitoring/inventory of PCBs in sealants: Switzerland

- **Switzerland did a nation-wide comprehensive screening of PCB in polymer sealants in buildings.**
- **1348 samples of joint sealants** from concrete buildings built between 1950 and 1980 were **analyzed**. **568 samples (42%) exceeded the limit of 50 mg PCB/kg.** PCB concentrations were **above 10,000 mg/kg in 21% of samples**, and **above 100,000 mg/kg in 9.6% of samples.**
- The study revealed: • The use of PCBs in joint sealants was a common construction practice in Switzerland between **1955 and 1975**. • The sealants represent a significant stockpile of PCBs.
- In a **subset of 85 samples,** it was investigated whether wavelength-dispersive X-ray fluorescence spectrometry (**WD-XRF**) can be used as a rapid screening method for joint sealants.
  - **Detection limit for PCBs** with **WD-XRF: 10 to 25 mg/kg. Other XRFs: 100 mg/kg (or lower)**
  - Detected chlorine concentrations were between <1 g/kg and 200 g/kg
  - However, chlorine sources other than PCBs, in particular chlorinated paraffins, other chlorinated organic compounds, and inorganic chlorine, need to be considered.
    - **Chlorinated paraffins were detected in about one-third of the samples of the subset.**
    - Therefore, low specificity of WD-XRF for PCBs. Since SCCPs/MCCPs are POPs/POP candidates their detection is also appreciated today.





Kohler et al., Environ. Sci. Technol. 2005, 39, 1967-1973

Monitoring/inventory of PCBs in sealants: Canada

- In Canada in ca. 20% of buildings constructed between 1945 to 1969, PCBs were detected in sealants in a monitoring study.
- In particular institutional buildings (school, university) and commercial buildings affected.
- In simple residential buildings no PCBs in sealants were detected in Canada.

Building age	Number of buildings sampled	Number of buildings with PCB-containing sealants	% Detection
Pre-1945 (control)	8	0	0
1945–1960	11	3	27
1960–1969	41	7	17
1970–1980	28	1	4
Post–1980 (control)	21	0	0
Building type			
Institutional buildings	32	8	25
Commercial buildings	6	1	17
Residential buildings	34	0	0
Industrial buildings	3	0	0
Infrastructure	5	1	20
Single-detached residential	13	1	8
Construction material			
Concrete buildings	45	9	20
Brick buildings	32	1	3
Other (glass, stucco, stone, etc.)	32	1	3

Inventory of PCBs/PCNs and SCCPs in Open Application: **Sealants and Adhesives in South Africa**

Inventory of PCBs/PCNs/SCCPs in sealants and adhesives:

- **South Africa had several major constructions in the 1950s to 1970s** in the time where PCBs and possibly PCNs were used as additives in sealants.
- This includes the construction of dams which for Europe has been reported to contain PCBs in sealants and paints: Kougha Dam (1957-1969), the Gariep Dam (1971 originally named the Hendrik Verwoerd Dam), Sterkfontein Dam Reservoir (construction start 1969 and commissioned 1977), Vanderkloof Dam (1973-1977) and Kilburn Dam (1970s opened 1981).
- Also dams and hydropower stations were constructed in the **1980s and 1990s** (e.g. Colley Wobbles Power Station is a hydroelectric power facility; Bethlehem Hydro). These do likely not contain PCBs or PCNs **but might contain SCCPs in sealants and paints**.
- Assessment needs on the content and current approach in repair. Such paints or sealants might be removed with sand blasting with associated contamination of the river/lake and wider environment. This practice need to be prohibited.

Weber R, Okonkwo J (2019) Assessment and Preliminary Inventory (Tier 1 and 2) of PCNs, SCCPs and PCBs1 in South Africa. Report for the BRS Secretariat.

Inventory of PCBs/PCNs and SCCPs in Open Application: **Sealants and Adhesives in South Africa**

Inventory of PCBs, PCNs and SCCPs in sealants and adhesives:

- Experience in Europe/US show that university and school buildings built in the 1950s to 1970s can contain PCBs.
- **Large (prefabricated) concrete buildings were constructed in South Africa.** This might concern: University of KwaZulu Natal (1960), Zululand Univ. (1960), Nelson Mandela Metropolitan Univ. (1964), Univ. of Johannesburg (1966), Port Elisabeth Technikon (1967) Vaal Uni. of Technology (1967); Tshwane Univ. of Technology (1967s), Cape Peninsula Univ. of Technology (1967), Durban Univ. of Technology (1967), ML Sultan Technikon (1969) , Peninsula Technikon (1972) Sefako Univ. of Medical Health Medunsa (1976), Walter Sisulu Univ. (1977), Mangosuthu Univ. of Technology (1979) (Cooper & Subotsky 2001).
- These buildings might contain sealants with PCBs or PCNs but also SCCPs that have already been produced at that time. Screening task!

Weber R, Okonkwo J (2019) Assessment and Preliminary Inventory (Tier 1 and 2) of PCNs, SCCPs and PCBs1 in South Africa. Report for the BRS Secretariat.

Inventory of PCBs/PCNs and SCCPs in Open Application: **Sealants and Adhesives in South Africa**

Survey in companies producing or importing sealants & adhesives:

- An initial screening of companies producing or importing sealants has been conducted.
- South Africa has a National Adhesives & Sealant Manufacturing Association (N.A.S.M.A.; founded in 1975) to promote & develop the adhesive & sealant industry in South Africa. The association consist of members who are responsible for 75% of adhesives sold in South Africa. Sealant providers include e.g. Adtec, Alcolin, BASF, Builders, Den Braven, Henkel, Pattex, Permoseal, Qualichem-Genkem.
- The overall production portfolio of these companies covers all sealant types which possibly contain SCCPs (and formerly might have contained PCBs/PCNs) such as polysulphide sealants, acrylic sealants, butyl sealants, or polyurethane sealants.

Assessment need for their current and former portfolio.

Weber R, Okonkwo J (2019) Assessment and Preliminary Inventory (Tier 1 and 2) of PCNs, SCCPs and PCBs1 in South Africa. Report for the BRS Secretariat.

Overall use/inventory of PCBs in open applications: Germany

Approx. 25,000 t of PCB have been used in Germany in open applications from 1950s to early 1970s.

- 20,000 t PCB were used as plasticizer in sealants (Detzel et al. 1998). Major use in public buildings (e.g. kindergardens, schools, university and office buildings). But also in industry and residential buildings.
- 4000 t PCBs were used in paints and coatings in buildings (on walls, floors, ceiling tiles, electric cables) and constructions (swimming pools, bridges, pylons, silos, pipes), as lubricating/cutting oils and adhesives.





Pictures: Christine Herold, Urs Wagner, Roland Weber Michael Bührke/pixelio.de Jakob Ehrhardt/pixelio.de

PCBs in building materials in Germany

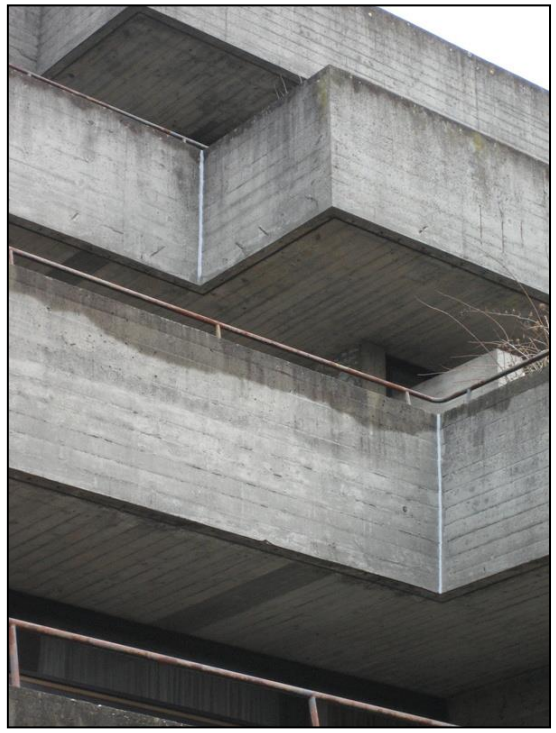
- **In 1973 PCB use in open applications have been restricted in Germany** and sales have stopped. Stockpiles of PCBs might have been used from construction companies also after the restriction of use to finish these **stockpiles (maybe until 1975).**
- This information is important for inventory development.

- Sealants and industrial paints have long service life of 30 to more than 60 years and are now becoming increasingly relevant in building demolition and in renovation projects.
- As a result, large amounts of PCBs remain present in buildings until today. For Germany it was estimated that 50 to 80% of sealants in buildings and constructions were still in use in 2013.
- In Denmark where the largest share of sealants were used for window sealants, a lower share of PCBs in sealants were considered still present.
- The management problems of PCB sealants are growing when the buildings and constructions get older, because maintenance, repair and renovation work is carried out more frequently.

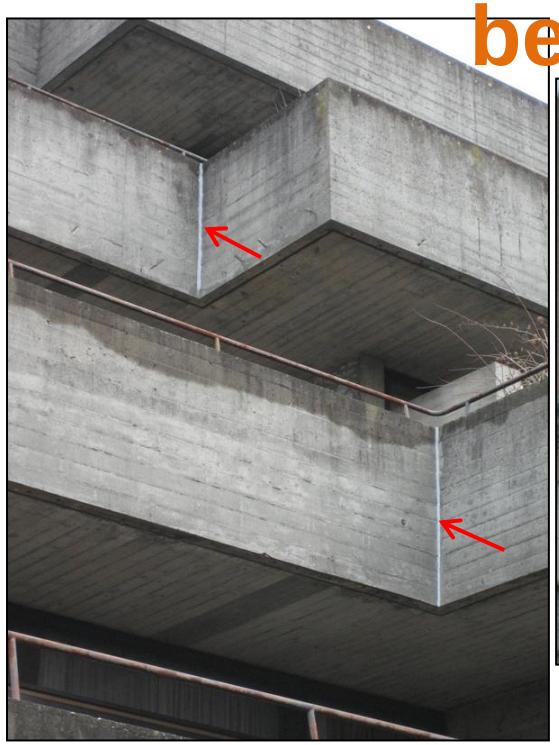
PCB use in sealants

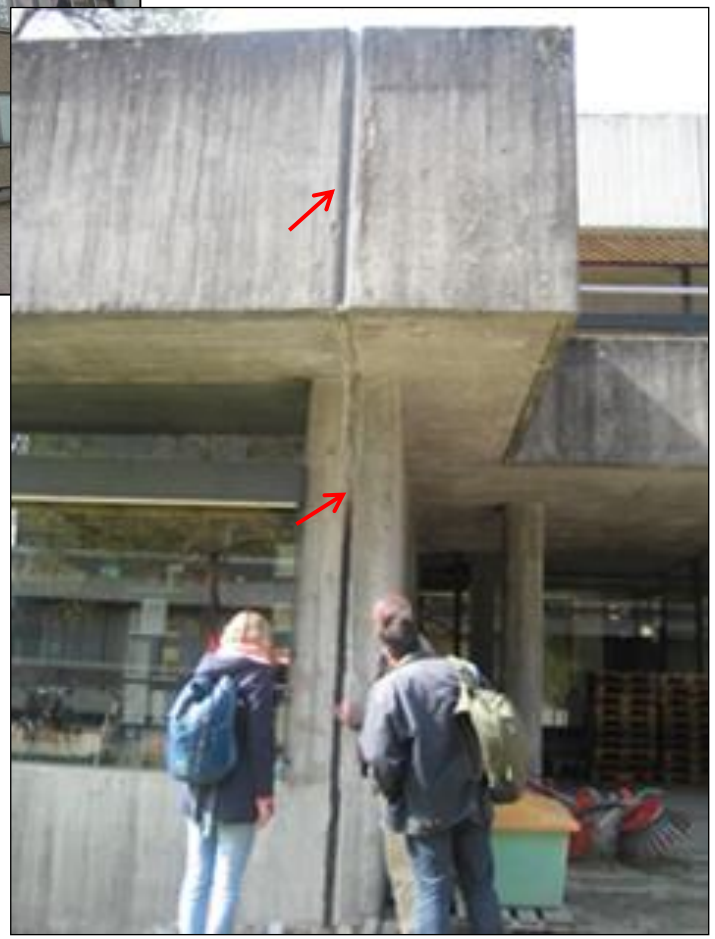
PCBs have been mainly used:

- In outdoor sealants in seams between concrete blocks.
- In indoor elastic sealant materials
- In elastic sealants around windows and door frames

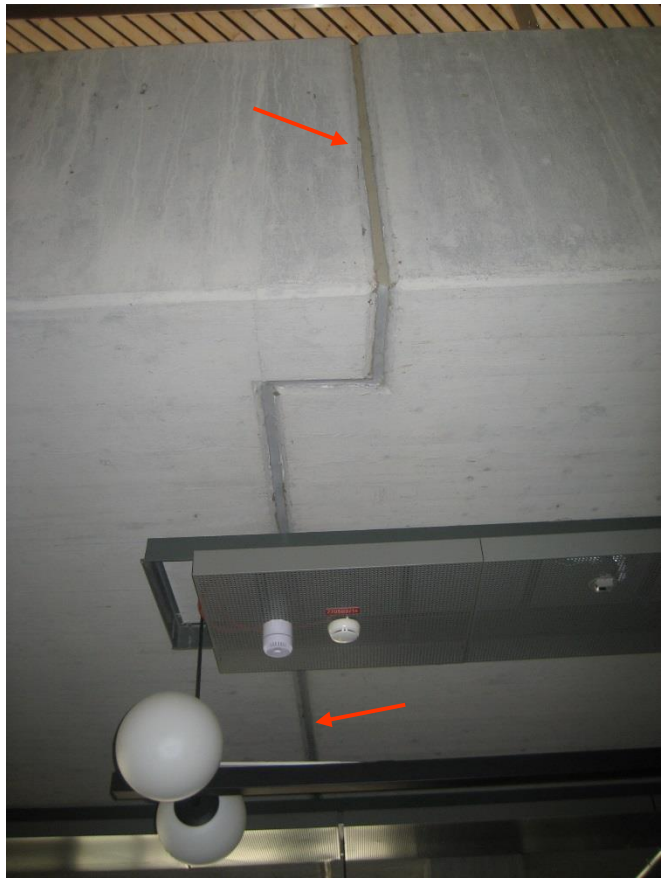


Examples of PCBs in outdoor sealants between concrete blocks



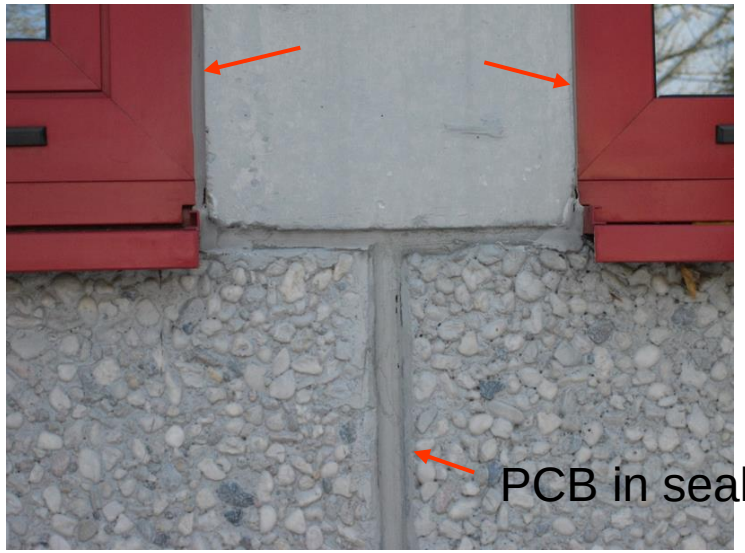


Examples of PCBs in indoor elastic sealant/caulk materials



Examples of PCB-containing sealants around doors and window frames

PCBs in a narrow strip of sealant between window frame and windowsill



PCB in sealant between concrete blocks

PCBs in sealants of double-glazed windows (Denmark & Norway)

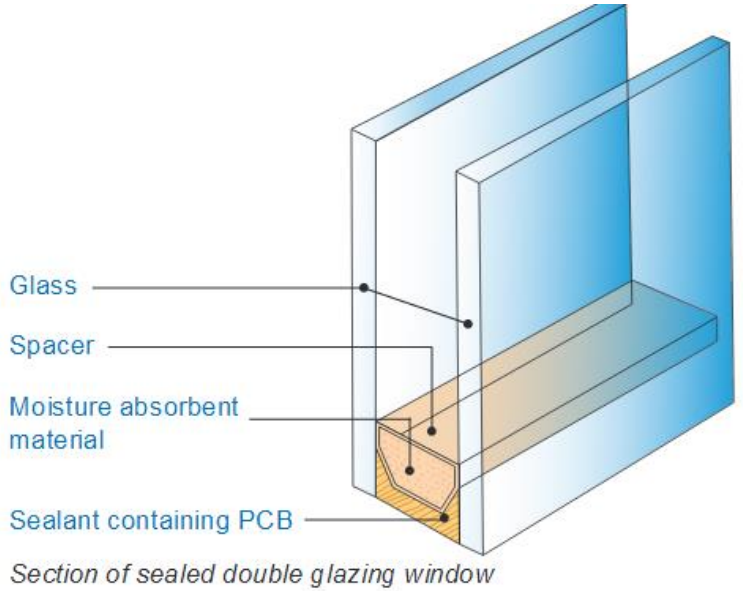
Denmark

•In Copenhagen University Hospital some of the outside face joints and butyl seals between glass bead and window glass in the double glazing contained varying concentrations of PCB.

http://www.alectia.com/en/projects/pcb-mapping-at-copenhagen-university-hospital-en/

Norway

- More than 2 million sealed double glazing windows containing PCB have been used in buildings in Norway.
- The sealing adhesive contains around 70 grams of PCB per window.



Section of sealed double glazing window

http://www.ruteretur.no/file/Folder%20engelsk%202012.pdf

STOCKHOLM CONVENTION

PCBs in Open Applications:
PCBs in paints and anti-corrosion coating

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PCB use in paints and coatings

- In several countries PCBs have been found in surface coatings like paint or plaster (Canada, Denmark, Germany, Norway, Sweden, Switzerland, USA). **Information for most other countries is missing.**

- **PCBs were popular plasticizers for paints in the 1950s to 1970s**. Up to 25 % PCB was added to some formulas.
- Also PCNs have been used in paints from 1930s to 1980s).
- Chlorinated paraffins are still used today in paints and coatings.

- In 1973 OECD recommended restrictions on the use of PCBs, and PCBs in paints were gradually replaced by chlorinated paraffins (and possibly others).

Jartun et al. (2009) Environmental Pollution 157, 295–302
Johnsen and Engoy (1999) Contamination from marine paints https://apps.dtic.mil/sti/citations/ADP010602

PCB use in paints and coatings

- Paints are mixtures mainly consisting of: binder, liquid, pigment, and additives.
- The binder is a polymer or a resin, which provides the basis of the continuous paint film by adhering the pigment particles together.
- PCBs were added to the binder as a plasticizer
- **Main paint/coating types where PCB were used:**
  - Chlorinated rubber paint/coating. Chlorinated rubber paint has great resistance to acids, alkalis, salt solutions and water;
  - PVC copolymer
  - Epoxy paint; and
  - Vinyl acetate-paint.
- **PCBs gave the paint good adhesive properties and resistance in paints/coating against moisture, chemicals, corrosion, and flames.**

Jartun et al. (2009) Environmental Pollution 157, 295–302
Johnsen and Engoy (1999) Contamination from marine paints https://apps.dtic.mil/sti/citations/ADP010602

PCB in paints and other surface coatings

PCBs were used in

- **Corrosion resistant paint/coating on metal constructions** (mainly chlorinated rubber and PVC-copolymer coatings),
- paints on walls, floors, and facades,
- plaster on walls and facades,
- coating of slip-resistant floors,
- paints on doors and window frames,
- varnishes, lacquers,
- waterproofing coatings,
- mold resistant coatings,
- flame retardant coatings,
- **road line paint.**

BUWAL (2000)
Jartun et al. (2009) Environmental Pollution 157, 295–302
Johnsen and Engoy (1999) Contamination from marine paints https://apps.dtic.mil/sti/citations/ADP010602

PCBs in anti-corrosion coatings

Flaking anti-corrosion paint

Tank, anti-corrosion coating

UNEP: PCB IN OPEN APPLICATIONS: MACHINERY AND INSTALLATIONS (Picture ETI), http://chm.pops.int/Implementation/PCBs/DocumentsPublications/tabid/665

PCBs in anti-corrosion coatings

- In a range of outdoor anti-corrosion applications PCB paints have been verified including (metal) bridges or pylons.
- The share of PCB paints in anti corrosion resistant paints on metal constructions (mainly chlorinated rubber and PVC-copolymer coatings) is not known. Also alternative chlorinated organics were used (SCCP/MCCP; PCNs)
- For “dry” applications often heavy metal paints were used (lead, zinc).

Pylons

Bridges

Ships and airplanes

ETI Umwelttechnik AG

UNEP: PCB IN OPEN APPLICATIONS: MACHINERY AND INSTALLATIONS; (Pictures ETI), http://chm.pops.int/Implementation/PCBs/DocumentsPublications/tabid/665

PCBs in anti-corrosion coatings in machinery and installations

Turbines and generators

Pumps and motors

Pipelines

Cranes

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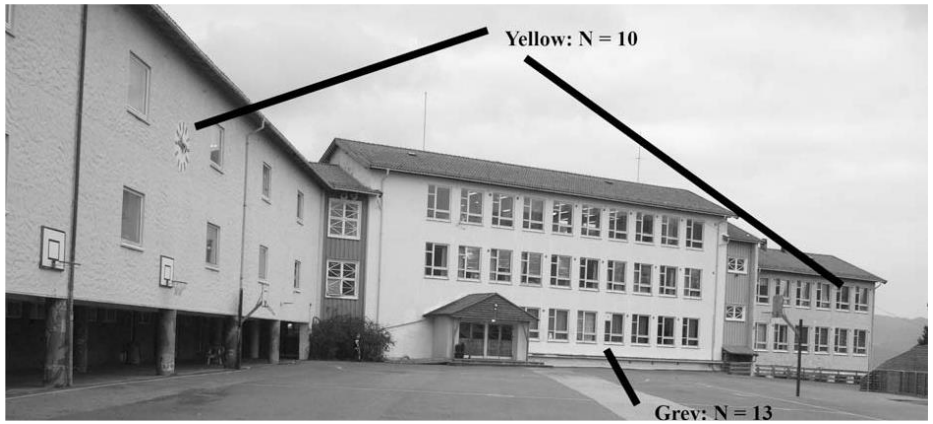
UNEP: PCB IN OPEN APPLICATIONS: MACHINERY AND INSTALLATIONS; Pictures ETI, http://chm.pops.int/Implementation/PCBs/DocumentsPublications/tabid/665

PCBs in plaster and paint on walls & façades



Jartun et al. (2009) Environmental Pollution 157, 295–302

UNEP Photo Booklet on Open Applications
http://chm.pops.int/Implementation/PCBs/DocumentsPublications/tabid/665

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PCBs in paints on concrete, plaster and screed floors

Paint in the basement of an appartment building

Paint on concrete (floor, walls), paint on fire protection door

Paint in the laundry-room of an appartment building: Paint on screed (floor) and plaster (wall)

PCBs in paints on concrete, plaster and screed floors

Paint on floors

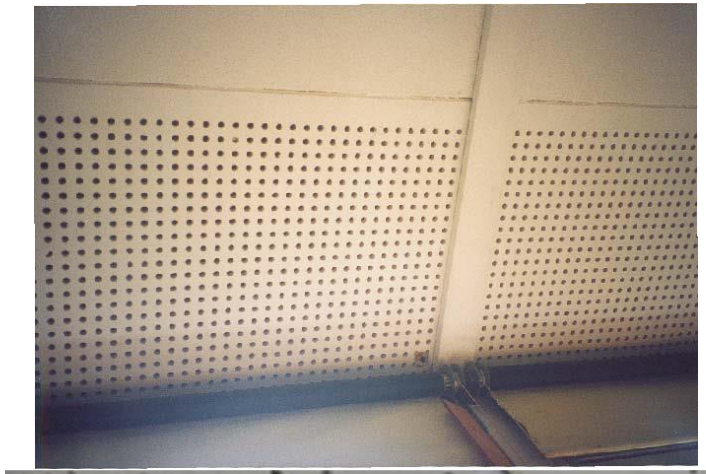
Paint and sealants on balconies

ETI Umwelttechnik AG

UNEP Factsheet on Open Applications: Residential and Public Buildings
http://chm.pops.int/Implementation/PCBs/DocumentsPublications/tabid/665

Acoustic ceiling tiles

PCB were used as flame retardants for wood and cardboard in indoor applications.



Guidebook NRW (2003) PCBs in buildings (in German)

UNEP Photo Booklet on Open Applications
http://chm.pops.int/Implementation/PCBs/DocumentsPublications/tabid/665

top: unpainted side; bottom: painted side

Guo et al. (2011) US-EPA, https://clu-in.org/download/contaminantfocus/pcb/PCB-lab-studies-1.pdf

PCB-containing paints in swimming pools

- Large use as coating in swimming pools.
- In Switzerland it es estimated that 20 % of outdoor swimming pools are contaminated.
- **Swiss inventory of swimming pools** developed by regional competent authorities.
- City Hannover/Germany screened their public pools for PCBs (inventory city level) and found some PCB contaminated.

- Because of inappropriate handling during previous renovations the surrounding ground around swimming pools is sometimes contaminated above the PCB limit value for soil.

Removal of PCB-containing paint in a swimming pool in Zurich (Switzerland)

http://www.pollubat.ch/de/section/links-pcb

PCBs in paint/coating on farms: silos, barns & manure pits

- PCB paints where used in silos as coatings to protect concrete/metal against acidic feed. This was the major source of PCB contamination of cow milk in Germany.
- PCB paint has been applied inside the barn and manure pit,
- This coating was one of materials in 50 to 70s strong enough to withstand gases arising from the manure (likely based on chlorinated rubber or PVC copolymer).
- The transfer of PCB in manure resulted in PCB contamination of agricultural areas.
- Also in this application substitution by chlorinated paraffins and probably

Bild: Michael Bührket/pixelio.de

Willett LB, Hess JF (1975) Polychlorinated biphenyl residues in silos in the United States. Residue Rev 55:135–14

Weber et al. (2018) Life cycle of PCBs and contamination of the environment and food from animal origin. Environ Sci Pollut Res Int. 25(17), 16325-16343 SCP/RAC publication

The inappropriate removal of paints may cause serious contamination

- Essential during paint removal is the enclosure of the working area. However, it had been found in Switzerland that 5 to 10 % of the coating of pylons got lost, even despite these protective measures.
- In 2000 the Swiss EPA developed a technical guideline to reduce PCB-emissions during removal of anti-corrosive paints (in German).

BAFU (Swiss EPA) (2000)

- In September 2016 high PCB-concentrations were found in a stream of a Swiss national park. Sandblasting of an anti-corrosive paint on a dam had released PCB-contaminated dust to the waterbody.

PCB-contamination caused by inappropriate removal of paints

Puddefjordsbroen Bridge (Bergen, Norway)

- Bridge close to the Norwegian coast
- Concrete bridge covered with plaster and a layer of PCB-containing paint when built in 1956

Alasdair McLellan (CC BY-SA 3.0)

- 11,000 m^2 painted surface area
- Total amount of PCBs: 1650 kg PCB.
- Completely sandblasted in the mid-1980s
- Marine sediments below the bridge are heavily contaminated with PCBs.

Jartun et al. (2009) Environmental Pollution 157, 295–302

PCB-contamination caused by inappropriate removal of paints

Photo: Drehscheibe-online, Bildeinsteller „jutei“,
Picture from 17.05.2015

- PCB contamination of the river Elbe is monitored since the 1990s.
- In spring 2015 a railway bridge with anti-corrosive coating was renovated in a Czech city.
- Despite enclosure of the working area (photo) approx. 500 kg PCB (100 kg PCB_6) ended up in the river Elbe.

Schwartz et al. (2015)
https://www.researchgate.net/publication/308016270_PCB_in_River_Elbe_-_characteristics_occurrence_and_trends_as_well_as_causes_and_effects_of_increased_release_in_2015

PCB-contamination caused by inappropriate removal of paints

Photo: Drehscheibe-online, Bildeinsteller „jutei“, Aufnahme vom 17.05.2015

- PCB-contaminated sediments are moving towards the Port of Hamburg (distance of 500 km).
- Here every year ca 3 million cubic meters of sediments have to be removed from the waterway to secure access for seagoing vessels.
- PCB-contaminated sediments are not allowed to be dislocated.

- Shipping traffic in the biggest German harbor is at risk to stop if the contaminated sediments are not removed in the upstream area.

Schwartz et al. (2015)
https://www.researchgate.net/publication/308016270_PCB_in_River_Elbe_-_characteristics_occurrence_and_trends_as_well_as_causes_and_effects_of_increased_release_in_2015

Contamination of harbour sediments through PCBs in paint of ships

- Chlorinated rubber was used as a binder in marine paint until the early 1990s. PCBs were added to chlorinated rubber as a plasticizer in a concentration of about 10 % resulting in a total concentration of PCB in paints of about 2 %.
- Average 2000 liters of PCB-containing paints were used on a ship.
- Paint is lost during painting and cleaning (sandblasting and other cleaning methods).
- It is likely that marine paints are responsible for most of the historic PCB contamination of harbour sediments in Norway. (But for Oslofjord for example, run-off from urban surfaces is the most important source today).

Johnsen and Engoy (1999) Contamination from marine paints https://apps.dtic.mil/sti/citations/ADP010602

Norwegian Environment Agency (2014) Contaminants in coastal waters of Norway 2013, https://www.miljodirektoratet.no/globalassets/publikasjoner/m250/m250.pdf

Secondary contamination in buildings from PCBs in sealants/paints

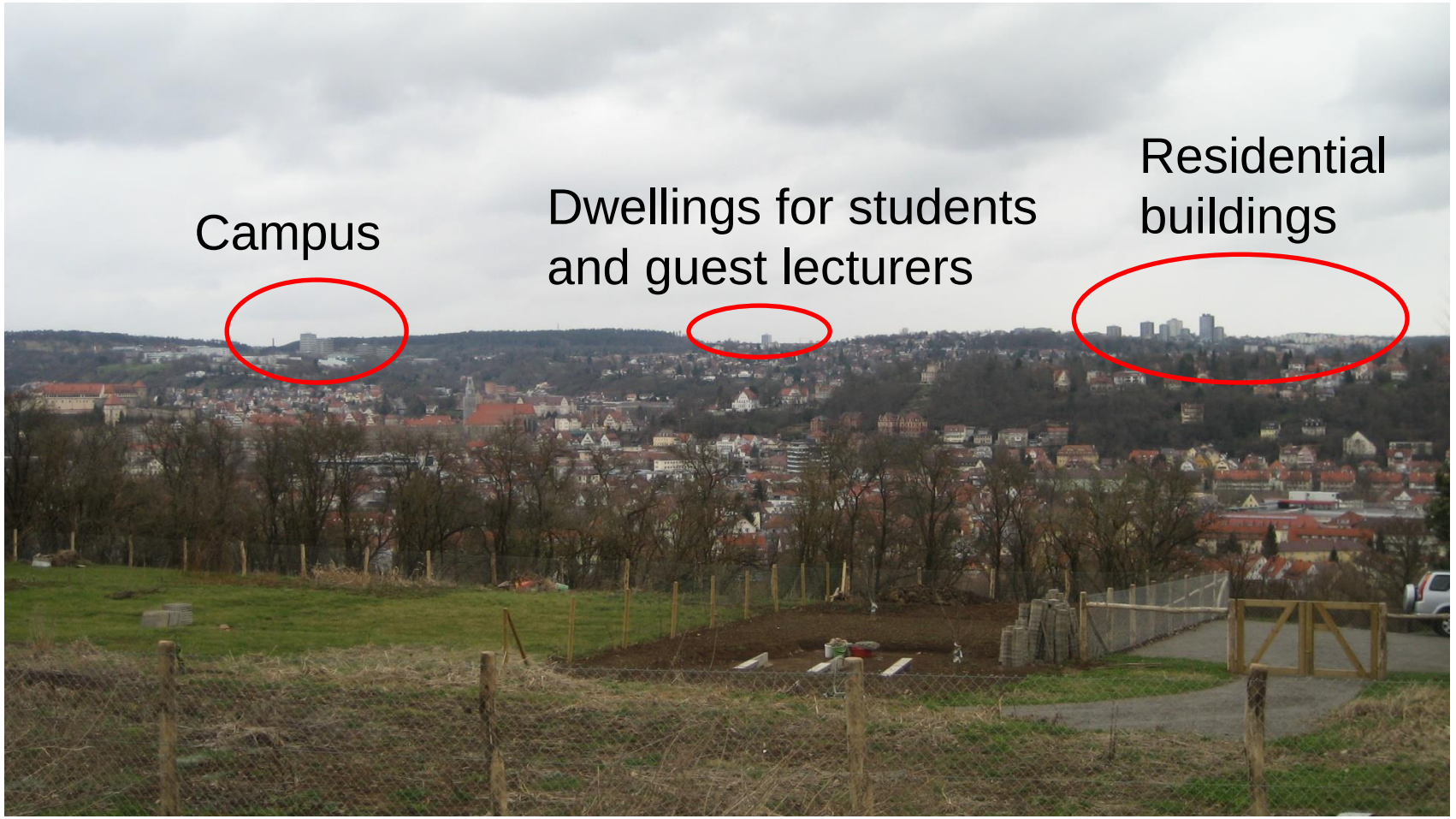
Secondary PCB contamination and resulting secondary sources

- PCBs in the air are absorbed into other building materials, furnishing and all other objects in the room (e.g. books, clothing), and dust.
- After some years of exposure those materials can reach high PCB-concentrations, sometimes above the limit for POPs waste of 50 mg PCB/kg.
- Case Switzerland: School with PCB sealants wanted to donate furniture (tables, chairs) to developing countries. The PCB level in furniture
- Because of its large surface area, dust can reach high concentration in a short time. In some buildings PCB levels in dust are > 50 mg PCB/kg and would be POPs waste.
- Secondary contaminated materials emit PCB themselves (secondary sources) which can become the main source after time and after renovation.

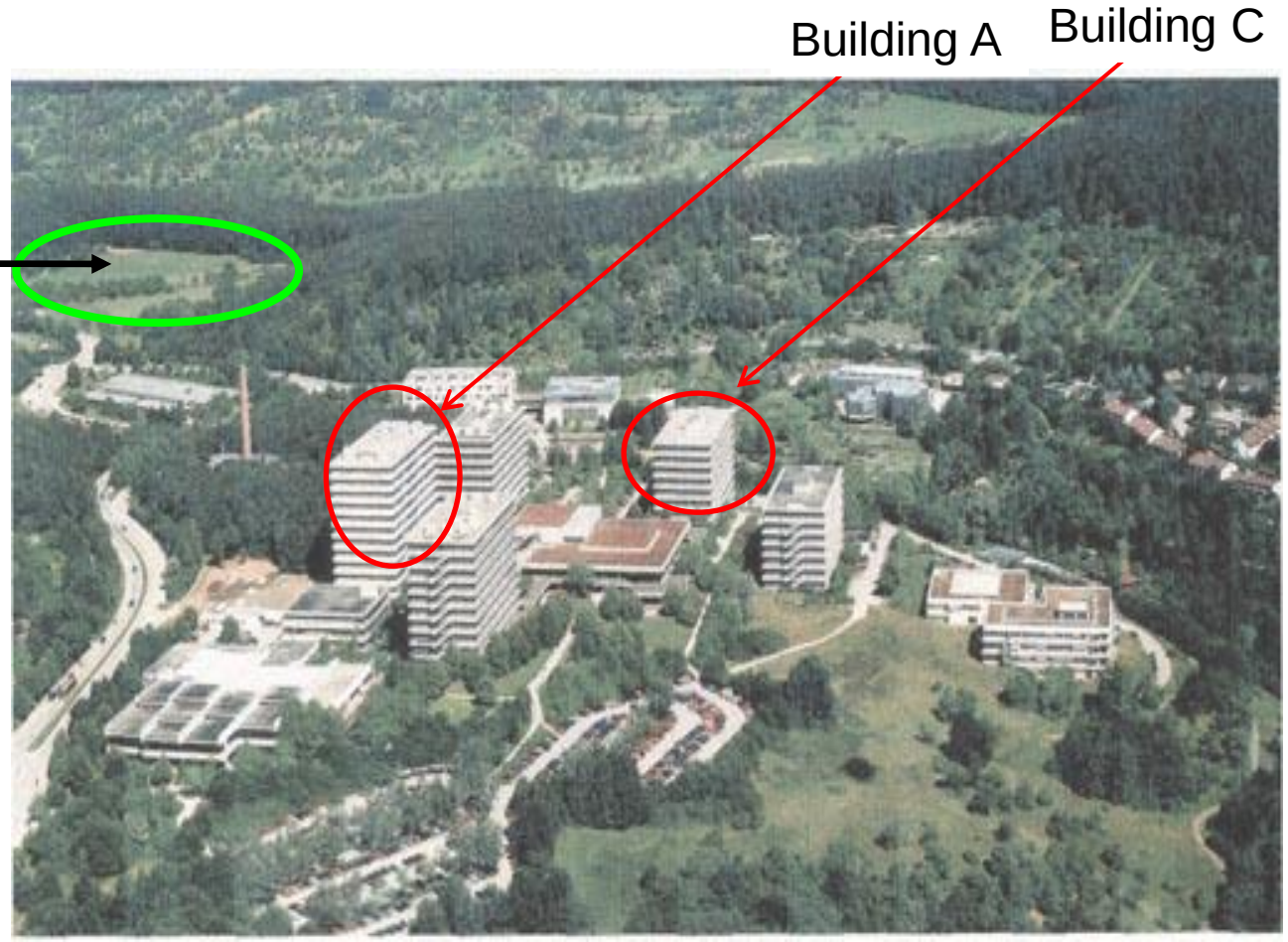
PCBs in building materials in Germany

Case study: Northern district of Tübingen/Germany

- Prefabricated buidlings and city areas which were constructed in the 1960s and 1970s are the major stock of PCB sealants and would be assessed in an inventory.
- The Northern part of Tübingen city is such an area which have been assesed to some extent.



PCB emission source: PCB in sealants and ceiling tiles on the campus Morgenstelle of the University of Tuebingen



- PCB-containing sealants in most of the buildings, indoors and outdoors.
- Ceiling tiles with PCB-containing paints in 3 buildings.
- PCB-containing paints on some doors and floors.
- PCB abatement only indoors – sealants on the facade remained!

Campus Morgenstelle of the University of Tuebingen
PCB abatement in building C (mathematics/physics)

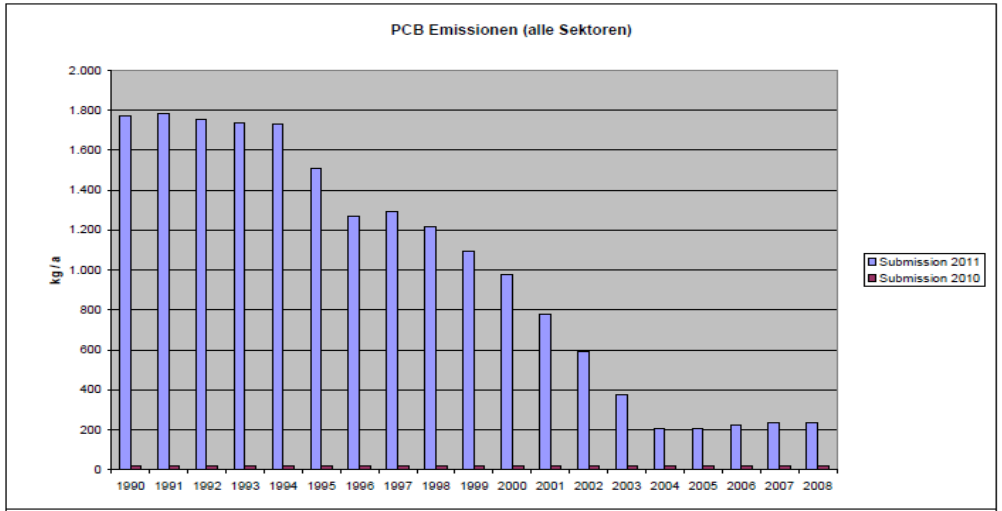
PCB inventory of the building C, prior to PCB abatement in 2005/06:

More than 1 t PCBs in sealants, ceiling tiles and paint.

- PCB emission through ventilation of indoor air prior to remediation (calculated with measured air exchange rate of 0.5/h, volume of the building and mean indoor air concentration 2.5 µg/m^3):
- Emission: ca. **600 g PCB/year**. This is six-fold above the threshold where facilities are required to report PCB emissions to the European Pollutant Release & Transfer Register (E-PRTR).
- **Annual emission rate: 0.06% of the mass inventory**
- This is comparable to the result of a Swedish study: emission rate of 0.067% per year from sealants (Sundahl et al.1999).

Total emission inventory of PCB amount & release building

- Assuming an annual emission rate of 0.06% and assuming that 50 to 80% of the 24,000 t PCB were still in use in Germany, this result in an emission of 5-15 t PCB per year for Germany.
- The calculated emission is in the same range as the annual emissions of 1.6 t PCB that Bogdal et al. (2014) extrapolated for Switzerland: Considering the different number of inhabitants an emission of 1.6 t /year for Switzerland would correspond to an emission of 16 t/year for Germany
- German unintentional PCB-emission inventory of thermal sources: 220 kg PCB/year.



PCB deposition/immissions in Germany

- Measured depositions in South Germany/Bavaria[3] background was (0,2 µg PCB/(m^2*d)) which would correspond to a **deposition of ca. 50 t** per year for West Germany and is therefore in the same order of magnitude as the estimated emission from open applications but two orders of magintude higher than the official PCB emission inventory.

⇒**The most relevant current PCB-sources for atmospheric deposition in Germany are open applications such as sealants & paints (PCB in closed applications managed finished around 2010 as EU-required). The higher deposition (50 tonnes) compared to emission estimate might result from additional releases during demolition or by remoblization from soils/environmental matrices.**

- The emissions from open applications make cities to PCB sources (see also studies in Zürich, Toronto or Birmingham). Emissions deposit in city & surrounding.

[1] Sundahl et al. (1999), J. Environ. Monit., [2] Weber/Herold estimate for a University building in Germany; [3] LfU Bayern (2006), [4] Csiszar (2012), PhD Thesis, [5] Csiszar et al. (2013), Environ. Sci. Technol., [6]Gasic et al. (2008), Environ. Sci. Technol., [7]Jamshidi et al. (2007), Environ. Sci. Technol.,

Cities as sources of PCBs to the wider environment - open application in buildings as source

Data from several other monitoring networks demonstrate elevated atmospheric PCB concentrations in urban areas

- For Toronto (Canada) estimated PCB stocks are 440 t and emissions are 230 kg PCB per year. The urban air plume was found to reach about 50 km (Csiszar et al. 2013, 2014)
- For Switzerland, emission estimate of PCB release per year was 1.5 t (Bogdal 2014)
- For Germany estimated PCB stocks in construction in 2013 were 12,000 to 19,000 t and the related emissions estimated to 5 to 15 t PCB per year by desorption without considering the impact of abrasive blasting! (Zimmermann et al. 2019).

Zimmermann (2019) UBA Texte 01/2019. Forschungskennzahl 3717 51 1010.

Other open PCB applications

There are some further open PCB applications with minor relevance

- Ingredient in sealants and caulking material
- Ingredient in paints and other coatings
- Plasticizer in polyvinyl chloride, cloroprene and other resins

- Ingredient of adhesives (can be relevant and can be assessed in buildings)
- Specific concrete admixture (Norway)
- Paper (short service life)
- Ingredient in inks (short service life)
- Ingredient in lubricants (short service life)
- Pesticide extenders/diluents (short service life)
- Lubricants for microscopes, brake linings, etc. (minor use)
- Flame retardant in fabrics, carpets, polyurethane foam
- Insulating materials of oil-filled power cables.

Inventory of remaining PCBs in Norway (2005)

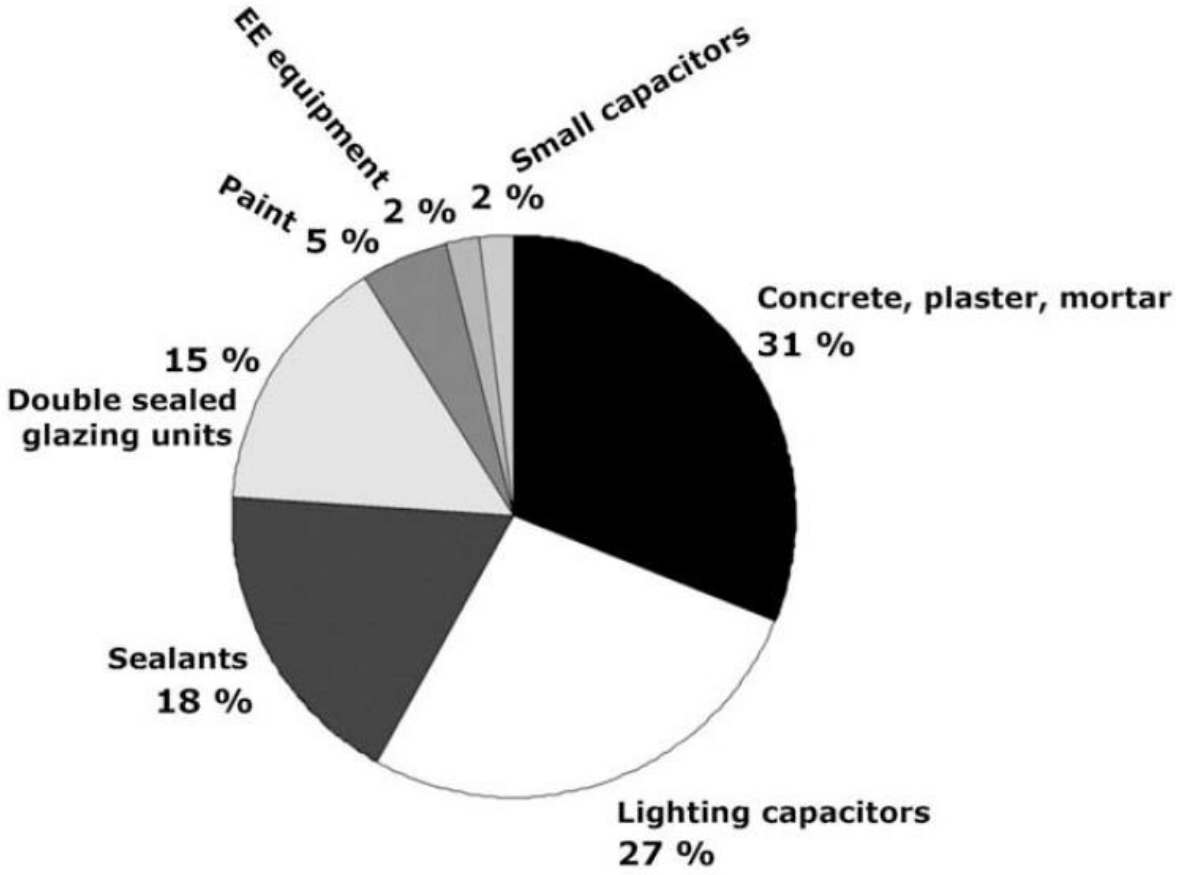


Fig. 1. Remaining polychlorinated biphenyls (PCBs) in products from Norway, shown by percentage of 155 tonnes. Numbers are from BNL (2005) and Norwegian Pollution Control Authority.

Remaining PCBs in Norway in 2005 (155 t):

- Largest stock were sealants (33% including double sealed glazing windows)
- Specific concrete/plaster/mortar (31%)
- Small capacitors (lights; EEE; 29%)
- Paint (5%)

Jartun et al. (2009) Environmental Pollution 157, 295–302; BNL (Byggenæringens Landsforening, The Federation of Norwegian Construction Industries), 2005. Identifisering av PCB i norske bygg. (Identification of PCBs in buildings from Norway). Nasjonal handlingsplan for bygg- og anleggsavfall. BNL(in Norwegian).

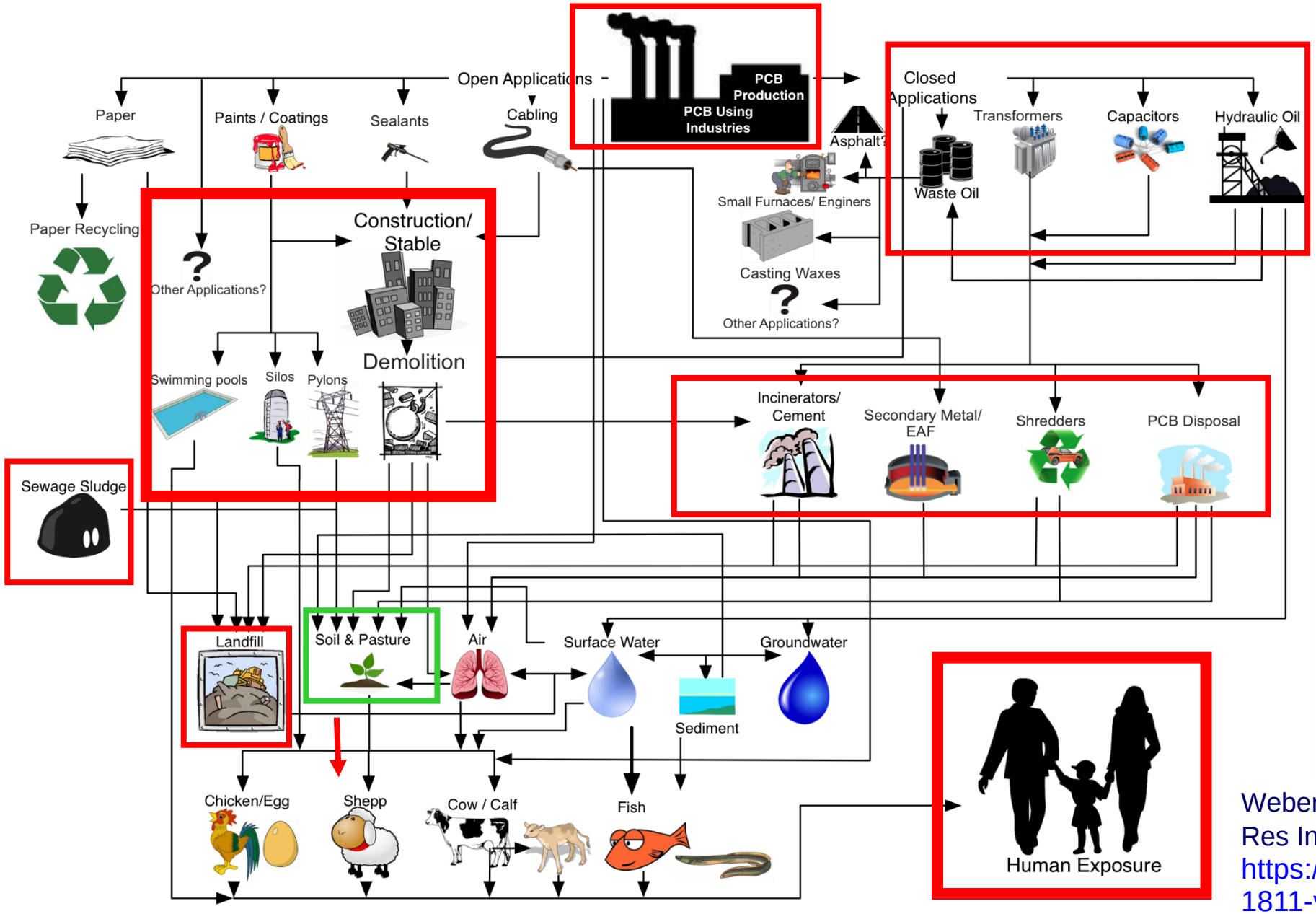
PCB use in nominally closed applications – hydraulic fluids

PCBs in hydraulic fluids was a major use in some countries in particular when having large mining activities.

- Hydraulic fluid use
  - **Mining equipment**
  - Machinery
  - **Military equipment**
  - Aluminium, copper, steel, and iron forming industries

Inventory Germany: Approximately **12,500 t PCBs (ca. 15% of total use)** were used in hydraulic fluids in Germany. **The major share was used in mining**. From these 12,500 t used in mining **only 200 t had documented ESM** at **end-of-life**. **Most hydraulic fluid were released during operation and then refilled**. Today the pumped water from mines in Germany are PCB contaminated with related environmental releases.

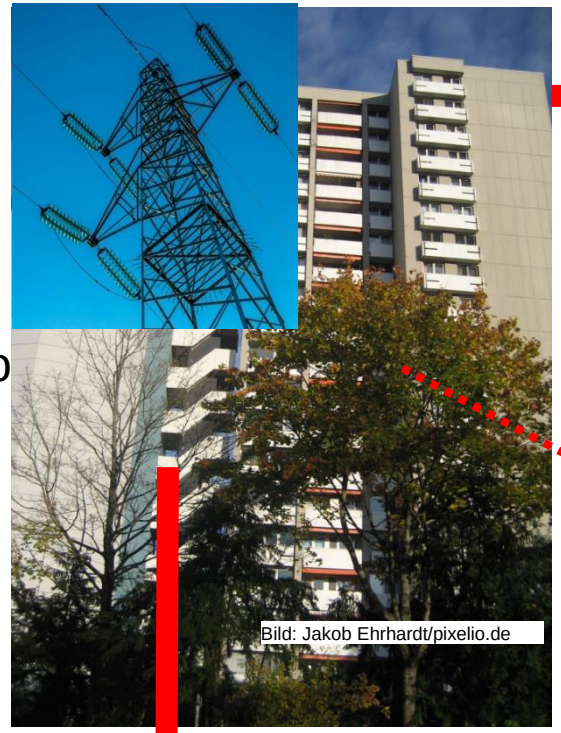
Open/closed PCB use, waste treatment and disposal/recycling, and related release and exposure pathways



Weber et al. (2018) Environ Sci Pollut Res Int. 25(17), 16325-16343 https://doi.org/10.1007/s11356-018-1811-y

PCB air emissions from open application and deposition on grass/feed and soils

Germany: 24000 t PCB in **open uses sealants & paints from 1960 to 1970**. Today still ~12000 t in use releasing 5 to 15 t of PCBs every year to the environment polluting soil and vegetation and livestock.



Exposure from soil

PCB-Exposure

PCB paints

Bild: Michael Bührket/pixelio.de

Direct exposure from point sources

Exposure from feed

Weber et al. (2018) Life cycle of PCBs and contamination of the environment and of food products from animal origin Environ Sci Pollut Res Int. 25(17), 16325-16343 https://doi.org/10.1007/s11356-018-1811-y

(dl-)PCBs in food animals in EU exceeding food limits

Dl-PCBs are a major reason that food animals exceeding EU maximum limits for total TEQ. Recent sources for exceedance of limits due to PCBs:

- PCB paints in silos for feed (silos constructed in the 1950s and 1970s)
- Wall paints in cattle stable
- Asbestos roof tiles painted with PCB paints.
- Paint on wood in chicken farm.

From Open Applications

- Rubber belt (from 1980s) in feeding trough for calves.
- Building with PCB sealants on pasture land
- Construction debris used for landscaping on pasture land

- Long term deposition from industrial facilities
- Former use as military area
- Area of a former railway line with railway sleepers
- Sediments from a dredged water reservoir with elevated PCB/PCDD/F
- Sewage sludge to agricultural soil in the 1960s/1970s
- Use of former scrap yard (PCB-contaminated) as storage area for dung.

From Contaminated sites

Germany: dl-PCBs in BIO eggs due to asbestos-cement fiber plates (ACP): Chemical and DR CALUX toxic dl-PCBs same!

Table 1
BEQ- and TEQ-values of PCDD/Fs and dl-PCBs found in three organic-farmed egg samples and one pooled chicken sample determined by a screening with the DR-CALUX® bioassay and confirmed by GC-HRMS.

DR CALUX = GC/HRMS

Sample	PCDD/Fs (pg TEQ/g)	dl-PCBs (pg TEQ/g)	Total TEQ (pg TEQ/g)	Total BEQ (pg BEQ/g)
Eggs 1	1.1	10.7	11.8	11.0
Eggs 2	0.72	5.7	6.4	6.2
Eggs 3	0.83	4.7	5.6	5.1
Chicken	0.57	10.6	11.2	11.0

Table 2
TEQ-values of dl-PCBs and PCDD/Fs and concentrations of non-dl-PCBs for eggs and soil samples according to their distances to the ACP-cladding.

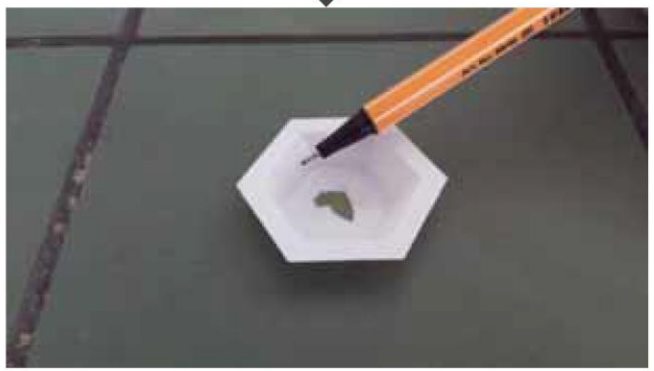
	Yard1[a]	Yard1	Yard2	Yard3	Yard4	Reference area
Distance to ACP (m)	0	0–1	2–8	1–5	4	35
Eggs						
dl-PCB-TEQ	–	10.7	5.7	5.4	2.4	–
Non-dl-PCBs	–	30,500	17,200	15,600	8700	–
PCDD/Fs-TEQ	–	1.1	0.6	0.7	0.5	–
Soil						
dl-PCB-TEQ	70.2	42.6	7.8	9.8	3.9	0.45
non-dl-PCBs	157,000	104,000	23,700	30,400	10,600	1200
PCDD/Fs-TEQ	3.5	3.8	1.5	1.8	2.1	1.3

Winkler (2015) High levels of dioxin-like PCBs in organic-farmed eggs: a case study. Env Intern 80, 72-78 (2015)

Wall paintings with high PCB content

- Several wall paintings with PCB contents of 3 to 16% (total PCB)
- Sum non-dl-PCB in the range of 1 to 6% → ΣPCB 101, 138, 153 and 180 corresponds 99% of the non-dl-PCB
- dl-PCB WHO-TEQ_{05} 200 ng/g → small chip (25mg) → 5 ng WHO-TEQ_{05}
- Application of the paintings more than 40 years ago

+1.5 mg non-dl-



Zennegg et al. 34th international Symposium of Halogenated POPs, 31. August- 5, Sept. Madrid, Spain

Monitoring POPs in C&D waste and recycling: PCBs

- PCB-sealants and paints can contaminate construction debris if not removed.
- Contamination of surroundings.
- Contamination of cattle when construction debris was used on farms for landscaping.
- **Recycling material is PCB-contaminated (PCB limit for recycling Germany 1 mg/kg Switzerland 0.5 mg/kg)**

Weber et al (2018) Environ Sci Pollut Res Int. 25, 16325-16343.

Indoor exposure in PCB contaminated buildings

Exposure from PCBs in buildings

- Open applications like sealants and paints emit PCBs directly into the environment and influence the PCB content in indoor air.
- PCBs in air are directly adsorbed by lung and skin.
- There is increasing evidence that in PCB-contaminated buildings inhalation of vapor-phase PCBs may for affected population be more important than ingestion via food.
- Residents of a flat with high concentrations of PCBs in the indoor air (about 1 µg/m3) were found to have four times higher levels of 27 congeners in the serum than unexposed controls (Meyer et al. 2013).
- Therefore if a country has discovered PCB in sealants, paints and small capacitors in buildings then further assessment might include human exposure studies.

Meyer et al. (2013) International journal of hygiene and environmental health 216.6, 755-762.

PCB stocks in open applications should be managed

- PCB in sealants and paints in large buildings/constructions can contain several 100 kg PCBs.

PCBs in sealants and paints should be inventoried and managed:

- Because these sealants are subject to the Stockholm Convention obligations for phase-out and disposal of PCBs, they should be classified and treated as PCB-contaminated material.
- Because of the relatively high levels of human exposure and their associated health effects compared to closed systems.
- Because open sources of PCB are continually exposed to a multitude of potential loss processes, such as
  - volatilization, wash-off and erosion.
  - (small) renovation, repair, maintenance and cleaning
  - or major renovation work

Some Conclusions on potential relevance of sealants/paints

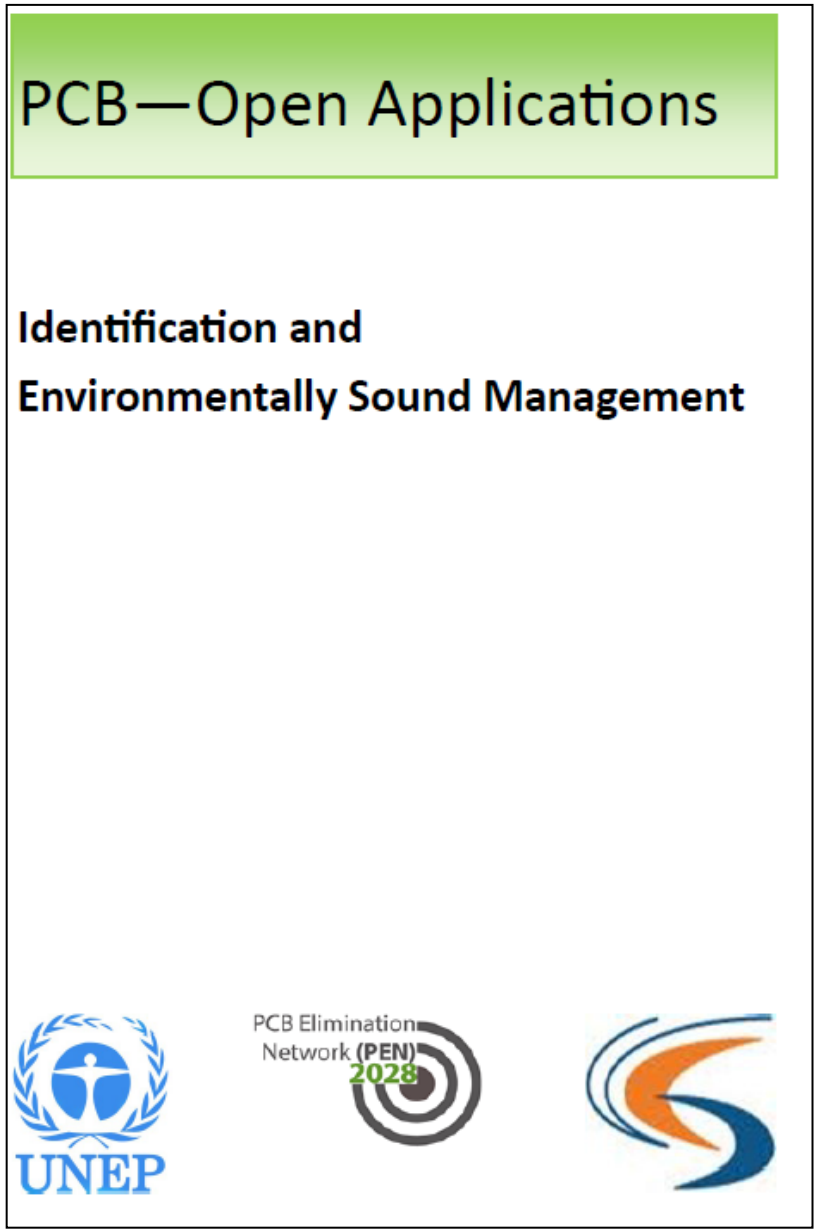
- Sealants are relevant current PCB sources in countries with former PCB use in this open application. Variation between countries ⇒ assessment need (Step 1)!
- Indoor exposure from PCBs in open applications is relevant. Difficult to achieve $TDI_{WHO(2003)}$ in PCB-building by renovation (due to secondary sources).
- Buildings constructed between 1950s to 1980 are often renovated with PCB releases from sealants and paints in particular when using sand-blasting with associated pollution.
- So blasting of paints can also release lead, cadmium or SCCPs/MCCPs depending on paints used in corrosion protection. Approach: addressing pollutants in paints in one inventory.
- Industrial countries still have challenges with management of sealants. Only Sweden has an inventory of PCB in open applications with a related systematic PCB management.
- Reductions of PCB concentrations in outdoor air and human exposure can be achieved by removing PCBs sealants in an environmentally sound manner from buildings and construction.

Some Conclusions on PCBs in sealants and paints/coating

- Sealants and paints are relevant current PCB sources in countries **with former** PCB paint application. Variation between countries! ⇒ assessment need!
- PCBs in sealants and paints in construction can enter sensitive material flows (recovery of C&D waste for landscaping) resulting in food exposure from recycling.
- PCBs in paints still relevant source for food exposure even in Germany which did some assessment of PCB paints in silos in the 1980s (but not a systematic assessment).
- Industrial countries have challenges with management of sealants and paints (and other OA uses like adhesives). Only Sweden has an inventory of PCB in buildings.
- Chlorinated paraffins (SCCPs, MCCPs) have substituted PCBs in paints/coating (SCCP listed as POPs in COP 2017 and MCCPs are proposed for listing possibly listed in 2025). Also lead was/is used in paints (SAICM emerging policy issue)

Information Materials on PCB in open application

ETI Umwelttechnik AG



http://chm.pops.int/Implementation/PCBs/Guidance/tabid/665/Default.aspx

Monitoring/management of POPs in construction and demolition waste

- PCB & **other POPs** should be monitored & managed when demolishing or renovating buildings.
- Monitoring of **POPs in buildings** (PCBs, SCCP/MCCP, HBCD, PBDE) need to be **integrated in** the **overall management of pollutants in buildings (e.g. asbestos, mineral fiber, lead).**
- Guidance documents on the assessment, deconstruction of buildings containing pollutants, were published e.g. by the Swiss and the Bavarian State Ministry (Germany).

Thank you for your attention !

More Information http://chm.pops.int/Implementation/PCBs/DocumentsPublications/tabid/665

Basel Convention: www.basel.int

Rotterdam Convention: www.pic.int

Stockholm Convention: http://chm.pops.int/

Montreal Protocol/Vienna Convention: http://ozone.unep.org

SAICM: http://www.saicm.org/

POPs phase out & alternatives http://poppub.bcrc.cn/

OECD/IOMC: http://www.oecd.org/chemicalsafety/

Science: www.ipcp.ch; http://greensciencepolicy.org/

NGO: www.ban.org; www.ipen.org; www.ihpa.info; www.chemsec.org

Better-world-links: http://www.betterworldlinks.org/