Polychlorinated Biphenyls (PCB)

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Introduction

Polychlorinated biphenyls (PCB) comprise a class of 209 individual compounds. The chemical structure and the numbering of the C-atoms are shown in Figure 1.

Figure 1: Structure and numbering for PCB

Sources and Chemical Identity of PCB

The two main sources of PCB are:

- Commercial production, and
- by-product in combustion processes as thermodynamically stable compounds.

PCB are produced by chlorination of biphenyl; its commercial production started about 60 years ago. The total amount produced world-wide is estimated at 1.5 million tons [Ivanov and Sandell 1992, Rantanen 1992].

Depending on the degree of chlorination of the PCB, their physico-chemical properties, like inflammability or electric conductivity, brought about a wide field of application. Thus, PCB have been used as electric fluids in transformers and capacitors, as pesticide extenders, adhesives, dedusting agents, cutting oils, flame retardants, heat transfer fluids, hydraulic lubricants, sealants, paints, and in carbonless copy paper. Some of their applications resulted in a direct or indirect release of PCB into the environment. Relatively large amounts were released due to inappropriate disposal practices, accidents and leakages from industrial facilities.

PCB were marketed with respect to percentage of their chlorine content (by weight) and were available under several trade names, *e.g.* Clophen (Bayer, Germany), Aroclor (Monsanto, USA), Kanechlor (Kanegafuchi, Japan), Santotherm (Mitsubishi, Japan), Phenoclor and Pyralene (Prodolec, France). In 1972, PCB production plants existed in Austria, Federal Republic of Germany, France, Great Britain, Italy, Japan, Spain, USSR, and USA. In the USA, the Monsanto industrial Chemical Company (the market leader with 98% of PCB/PCT production) terminated production and export in 1977. In Japan, the production of PCB was started by Kanegafuchi Chemical Co. Ltd. (Kaneka) in 1954 and marketed under the trade name "Kaneclor (KC)". PCB production, use and import were banned in Japan in 1972.

Production of PCB in Germany

In (West-) Germany the Bayer AG produced PCB under the trade name 'Clophen' (A30 to A60). In 1972, the Bayer AG restricted their supply of PCB for use in closed systems (transformers, condensers, hydraulic fluid). Until then approximately a total of 23,000 t of PCB were used in "open systems". In Table 1 the production of PCB in Germany for the years of 1974 to 1983 is listed separately for the various degrees of chlorination. It shows that there was shift in production from higher chlorinated PCB to lower chlorinated PCB over the years. It also shows that the use of PCB in Germany remained more or less constant up to 1978 and then decreased continuously. It is interesting to note that the production of PCB increased until 1980 with an increased proportion going into export. In 1983, the last year of PCB production by the Bayer AG, 90% of the PCB produced was exported.

Table 1: Production of PCB in West-Germany by degree of chlorination

t/year	Degree of Chlorination (% Cl)							Total Production	Used in Germany	Export
	39	42.5	47	48.5	54	55	60			
1974	--	2449	460	--	--	1619	1810	6338	2920	3258
1975		1648	292			1466	2141	5447	3400	2047
1976		2170	454			970	1436	5030	2789	2241
1977	139	2500	--	525	2516	--		5680	2910	2770
1978	690	4061		666	2223			7640	2824	4816
1979	937	3379		865	1963			7144	2446	4698
1980	799	4180		1127	1358			7464	2447	5017
1981	--	4778		--	--			4778	1180	3598
1982		3734						3734	968	2766
1983		4355						4355	430	3925

In Table 2 the distribution of PCB for the three closed systems transformers, condensers and coal mining are shown. While the use of PCB in condensers and transformers decreased continuously from 1974 to 1983 there was an increased use of PCB in mining. This was due to technical developments in coal mining with an increased demand for fire-resistant hydraulic fluids. Part of this demand had to be satisfied by import of PCB from the French company Prodelec. In coal mining new PCB was last used in Germany in 1985.

Table 2: Use of PCB in Germany 1974-1984 (t PCB)

Year	Condensers	Transformers	Coal Mining	Total
1974	1075	1130	871	3076
1975	752	1656	818	3226
1976	649	1125	930	2704
1977	637	740	967	2344
1978	446	590	1158	2194
1979	306	392	1361	2059
1980	253	334	1587	2174
1981	113	305	1350	1768
1982	30	318	1482	1830

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1983            6            40          1241        1287

1984            --           --          607         607

Sum 1974-1984   4267         6630        12372       23269
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Table 3: Nomenclature of PCB-homologues according to IUPAC [Ballschmiter and Zell 1980]

No.	Structure	No.	Structure	No.	Structure	No.	Structure
Monochlorobiphenyls		Tetrachlorobiphenyls		Pentachlorobiphenyls		Hexachlorobiphenyls	
1	2	52	2,2',5,5'	107	2,3,3',4',5	161	2,3,3',4,5',6
2	3	53	2,2',5,6'	108	2,3,3',4,5'	162	2,3,3',4,5,5'
3	4	54	2,2',6,6'	109	2,3,3',4,6	163	2,3,3',4,5,6
		55	2,3,3',4	110	2,3,3',4',6	164	2,3,3',4',5',6
Dichlorobiphenyls		56	2,3,3',4'	111	2,3,3',5,5'	165	2,3,3',5,5',6
4	2,2'	57	2,3,3',5	112	2,3,3',5,6	166	2,3,4,4',5,6
5	2,3	58	2,3,3',5'	113	2,3,3',5',6	167	2,3',4,4',5,5'
6	2,3'	59	2,3,3',6	114	2,3,4,4',5	168	2,3',4,4',5',6
7	2,4	60	2,3,4,4'	115	2,3,4,4',6	169	3,3',4,4',5,5'
8	2,4'	61	2,3,4,5	116	2,3,4,5,6		
9	2.5	62	2,3,4,6	117	2,3,4',5,6	Heptachlorobiphenyls	
10	2,6	63	2,3,4',5	118	2,3',4,4',5	170	2,2',3,3',4,4',5
11	3,3'	64	2,3,4',6	119	2,3,4,4',6	171	2,2',3,3',4,4',6
12	3,4	65	2,3,5,6	120	2,3',4,5,5'	172	2,2',3,3',4,5,5'
13	3,4'	66	2,3',4,4'	121	2,3',4,5',6	173	2,2',3,3',4,5,6
14	3,5	67	2,3',4,5	122	2',3,3',4,5	174	2,2',3,3',4,5,6'
15	4,4'	68	2,3',4,5'	123	2',3,4,4',5	175	2,2',3,3',4,5',6
		69	2,3',4,6	124	2',3,4,5,5'	176	2,2',3,3',4,6,6'
Trichlorobiphenyl		70	2,3',4',5	125	2',3,4,5,6'	177	2,2',3,3',4',5,6

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16 2,2',3	71 2,3,4',6	126 3,3',4,4',5	178 2,2',3,3',5,5',6
17 2,2',4	72 2,3',5,5'	127 3,3',4,5,5'	179 2,2',3,3',5,6,6'
18 2,2',5	73 2,3',5',6		180 2,2',3,4,4',5,5'
19 2,2',6	74 2,4,4',5	Hexachlorobiphenyls	181 2,2',3,4,4',5,6
20 2,3,3'	75 2,4,4',6	128 2,2',3,3',4,4'	182 2,2',3,4,4',5,6'
21 2,3,4	76 2',3,4,5	129 2,2',3,3',4,5	183 2,2',3,4,4',5',6
22 2,3,4'	77 3,3',4,4'	130 2,2',3,3',4,5'	184 2,2',3,4,4',6,6'
23 2,3,5	78 3,3',4,5	131 2,2',3,3',4,6	185 2,2',3,4,5,5',6
24 2,3,6	79 3,3',4,5'	132 2,2',3,3',4,6'	186 2,2',3,4,5,6,6'
25 2,3',4	80 3,3',5,5'	133 2,2',3,3',5,5'	187 2,2',3,4',5,5',6
26 2,3',5	81 3,4,4',5	134 2,2',3,3',5,6	188 2,2',3,4',5,6,6'
27 2,3',6		135 2,2',3,3',5,6'	189 2,3,3',4,4',5,5'
28 2,4,4'	Pentachlorobiphenyls	136 2,2',3,3',6,6'	190 2,3,3',4,4',5,6
29 2,4,5	82 2,2',3,3',4	137 2,2',3,4,4',5	191 2,3,3',4,4',5',6
30 2,4,6	83 2,2',3,3',5	138 2,2,3,4,4',5'	192 2,3,3',4,5,5',6
31 2,4',5	84 2,2',3,3',6	139 2,2',3,4,4',6	193 2,3,3',4',5,5',6
32 2,4',6	85 2,2',3,4,4'	140 2,2',3,4,4',6'	
33 2',3,4	86 2,2',3,4,5	141 2,2',3,4,5,5'	Octachlorobiphenyls
34 2',3,5	87 2,2',3,4,5'	142 2,2',3,4,5,6	194 2,2',3,3',4,4',5,5'
35 3,3',4	88 2,2',3,4,6	143 2,2',3,4,5,6'	195 2,2',3,3',4,4',5,6
36 3,3',5	89 2,2',3,4,6'	144 2,2',3,4,5',6	196 2,2',3,3',4,4',5',6
37 3,4,4'	90 2,2',3,4',5	145 2,2',3,4,6,6'	197 2,2',3,3',4,4',6,6'

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38 3,4,5	91 2,2',3,4',6	146 2,2',3,4',5,5'	198 2,2',3,3',4,5,5',6
39 3,4',5	92 2,2',3,5,5'	147 2,2',3,4',5,6	199 2,2,3,3',4',5,5',6
	93 2,2',3,5,6	148 2,2',3,4,5,6'	200 2,2',3,3',4,5,6,6'

Tetrachlorobiphenyls

	94 2,2',3,5,6'	149 2,2',3,4',5'6	201 2,2',3,3',4,5',6,6'
40 2,2',3,3'	95 2,2',3,5',6	150 2,2',3,4',6,6'	202 2,2',3,3',5,5',6,6'
41 2,2',3,4	96 2,2',3,6,6'	151 2,2',3,5,5',6	203 2,2',3,4,4',5,5',6
42 2,2',3,4'	97 2,2',3',4,5	152 2,2',3,5,6,6	204 2,2',3,4,4',5,6,6'
43 2,2',3,5	98 2,2',3,4,6	153 2,2',4,4',5,5'	205 2,3,3',4,4',5,5',6
44 2,2,3,5'	99 2,2',4,4',5	154 2,2',4,4,5,6'	
45 2,2',3,6	100 2,2',4,4',6	155 2,2',4,4',6,6	**Nonachlorobiphenyls**
46 2,2',3,6'	101 2,2',4,5,5'	156 2,3,3',4,4',5	206 2,2',3,3',4,4',5,5',6
47 2,2',4,4'	102 2,2',4,5,6'	157 2,3,3',4,4',5'	207 2,2',3,3',4,4',5,6,6'
48 2,2',4,5	103 2,2',4,5',6	158 2,3,3',4,4',6	208 2,2',3,3',4,5,5',6,6'
49 2,2',4,5'	104 2,2',4,6,6'	159 2,3,3',4,5,5'	
50 2,2',4,6	105 2,3,3',4,4'	160 2,3,4',4,5,6	**Decachlorobiphenyl**
51 2,2',4,6'	106 2,3,3',4,5		209 2,2',3,3',4,4',5,5',6,6'

As can be seen from Table 4, the number of possible isomers within the same degree of chlorination varies.

Table 4: Molecular formula, name, number of isomers, IUPAC number, molecular mass, percentage of chlorine and number of isomers identified

Molecular Formula	Name: Chlorobiphenyl	Number of Isomers	IUPAC-No.	Molecular Mass	% of Chlorine	No. of Isomers Identified

C12H9Cl	Mono	3	1-3	188.65	18.79	3
C12H8Cl2	Di	12	4-15	233.10	31.77	12
C12H7Cl3	Tri	24	16-39	257.54	41.30	23
C12H6Cl4	Tetra	42	40-81	291.99	48.65	41
C12H5Cl5	Penta	46	82-127	326.43	54.30	39
C12H4Cl6	Hexa	42	128-169	360.88	58.93	31
C12H3Cl7	Hepta	24	170-193	395.32	62.77	18
C12H2Cl8	Octa	12	194-205	429.77	65.98	11
C12HCl9	Nona	3	206-208	464.21	68.73	3
C12Cl10	Deca	1	209	498.66	71.10	1

Some physical and chemical properties of the PCB made them suitable for a broad range of applications. Important characteristics are:

- Liquid at room temperature; density: 1.1821.566 kg/L
- Low water solubility; readily soluble in organic solvents
- High flash point: 170-380°C,
- Non-explosive
- Low electric conductivity
- Very high thermal conductivity
- Extremely high thermal and chemical resistance (very high stability).

In general, melting point and lipophilicity increase with increasing degree of chlorination; vapour pressure and water solubility decrease. Thus, all PCB are lipophilic and poorly soluble in water. Water solubilities for Aroclors were determined in the range from 0.00270.42 ng/L.

Table 5: Water solubility for selected PCB (m/L)
a) Shiu & Mackay 1986, b) Dunnivant & Elzerman, 1988,
c) Weil *et al.*, 1974, d) Erickson, 1986

IUPAC-No.	S a	S b	S c	S d
0	5.94-7.48			
1	4.13-7.8		4.13	5.9
4	0.79-1.5	1.207	0.79	1.5
28	0.085-0.266	0.117	0.260	0.085
52	0.006-0.046	0.110	0.027	0.046

77	0.000569-0.175	0.00055	0.00075	0.175
101	0.00424-0.031	0.007	0.004	0.031
153	0.0012-0.0095			
182			0.00047	0.00048
209	0.000004-0.015			

Water solubility will increase in the presence of organic solvents (see Table 6).

Table 6: Differences in water solubility of PCB in pure water (S) and in water saturated with organic solvents (S*); expressed as log (S/S*)

IUPAC	Name	Methanol	Butanol	Octanol	Benzene	Benzyl alcohol
3	4-MonoCB	0.74	0.45	-0.10	-0.75	-0.12
30	2,4,6-TriCB	0.89	0.74	-0.26	-0.57	-0.04
61	2,3,4,5-TetraCB	1.02	1.25	-0.28	-0.77	-0.60
155	2,2',4,4',6,6'-HexaCB	1.22	1.76	-0.33	-0.70	0.39

The vapour pressure for some environmental pollutants are given in Table 7 and the vapour pressure from the solid phase and from the sub-cooled phase for selected PCB in Table 8. Compounds can be classified into three classes:

- Volatile organic compounds (VOC), *e.g.* acetone, dichloromethane, trichloroethane
- Semi-volatile organic compounds (SVOC), *e.g.* polycyclic aromatic compounds, chlorobenzenes, phthalates;
- Non-volatile organic compounds, *e.g.* polychlorinated biphenyls, polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans (depending on the degree of chlorination, they may belong to the class of SVOC).

Table 7: Typical ranges of vapour pressure for some environmental pollutants [Schwarzenbach *et al.* 1992]

Class of Compounds	Vapour Pressure at 25°C (atm)
Halogenated C1 and C2 hydrocarbons	10-2-1
Alkylbenzenes (butylbenzene-benzene)	5·10-4-10-1
Chlorobenzenes (hexachlorobenzene-monochlorobenzene)	10-8-5·10-2
Phthalate esters	10-7-10-4

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Polycyclic aromatic hydrocarbons (PAH)	10-11-10-3
Aliphatic hydrocarbons (C18-C5)	10-7-1
PCB	10-12-10-4

Table 8: Vapour pressure of selected PCB (atm); A= Dunnivant & Elzerman 1988; B= Murphy *et al.* 1987; C= Burkhard *et al.* 1984; D= Foreman & Bidleman 1985

IUPAC-No.	Substitution	A Solid	A Subcooled Liquid	B	C	D
4	2,2'	$1.82 \cdot 10^{-6}$	$4.1 \cdot 10^{-6}$	$1.5 \cdot 10^{-6}$	$4.18 \cdot 10^{-6}$	$3.31 \cdot 10^{-6}$
7	2,4	$1.79 \cdot 10^{-6}$	$9.86 \cdot 10^{-7}$	$1.73 \cdot 10^{-6}$	$2.09 \cdot 10^{-6}$	
9	2,5	$1.94 \cdot 10^{-6}$	$1.95 \cdot 10^{-6}$	$2.29 \cdot 10^{-6}$		
11	3,3'	$3.31 \cdot 10^{-7}$	$4.08 \cdot 10^{-7}$	$6.38 \cdot 10^{-7}$	$9.1 \cdot 10^{-7}$	
12	3,4	$7.26 \cdot 10^{-9}$	$1.27 \cdot 10^{-8}$	$5.25 \cdot 10^{-7}$	$7.75 \cdot 10^{-7}$	
15	4,4'	$3.24 \cdot 10^{-8}$	$5.41 \cdot 10^{-7}$	$5.01 \cdot 10^{-7}$	$7.4 \cdot 10^{-7}$	
18	2,2',5	$7.52 \cdot 10^{-7}$	$1.15 \cdot 10^{-6}$	$3.5 \cdot 10^{-7}$	$8.92 \cdot 10^{-7}$	$7.57 \cdot 10^{-7}$
26	2,3',5	$3.19 \cdot 10^{-7}$	$4.53 \cdot 10^{-7}$	$1.8 \cdot 10^{-7}$	$3.48 \cdot 10^{-7}$	$4.07 \cdot 10^{-7}$
28	2,4,4'	$1.43 \cdot 10^{-7}$	$3.0 \cdot 10^{-7}$	$1.5 \cdot 10^{-7}$	$2.73 \cdot 10^{-7}$	$3.31 \cdot 10^{-7}$
30	2,4,6	$6.36 \cdot 10^{-7}$	$1.5 \cdot 10^{-6}$	$9.34 \cdot 10^{-7}$	$1.09 \cdot 10^{-6}$	
40	2,2',3,3'	$1.08 \cdot 10^{-8}$	$9.44 \cdot 10^{-8}$	$4.5 \cdot 10^{-8}$	$1.1 \cdot 10^{-7}$	$8.69 \cdot 10^{-8}$
52	2,2',5,5'	$1.28 \cdot 10^{-7}$	$5.39 \cdot 10^{-7}$	$8.9 \cdot 10^{-7}$	$1.9 \cdot 10^{-7}$	$1.82 \cdot 10^{-7}$
53	2,2',5,6'	$6.62 \cdot 10^{-8}$	$4.0 \cdot 10^{-7}$	$1.1 \cdot 10^{-7}$	$3.51 \cdot 10^{-7}$	$2.63 \cdot 10^{-7}$
54	2,2',6,6'	$2.24 \cdot 10^{-8}$	$1.16 \cdot 10^{-6}$	$6.5 \cdot 10^{-7}$	$5.59 \cdot 10^{-7}$	
77	3,3',4,4'	$1.8 \cdot 10^{-10}$	$5.19 \cdot 10^{-9}$	$1.38 \cdot 10^{-8}$	$2.09 \cdot 10^{-8}$	
101	2,2',4,5,5'	$5.2 \cdot 10^{-9}$	$1.71 \cdot 10^{-8}$	$1.4 \cdot 10^{-8}$	$3.53 \cdot 10^{-8}$	$3.54 \cdot 10^{-8}$
104	2,2',4,6,6'	$4.28 \cdot 10^{-8}$		$1.68 \cdot 10^{-7}$	$4.28 \cdot 10^{-8}$	
128	2,2',3,3',4,4'	$2.9 \cdot 10^{-11}$	$4.6 \cdot 10^{-10}$	$0.97 \cdot 10^{-9}$	$3.54 \cdot 10^{-9}$	$3.62 \cdot 10^{-9}$

| 153 | 2,2'4,4',5, 5' | $3.2 \cdot 10^{-10}$ | $1.9 \cdot 10^{-9}$ | $2.5 \cdot 10^{-9}$ | $6.54 \cdot 10^{-8}$ | $6.91 \cdot 10^{-8}$ |
| 155 | 2,2',4,4',6,6' | $4.74 \cdot 10^{-9}$ | $3.49 \cdot 10^{-9}$ | $4.37 \cdot 10^{-8}$ | | |

Henry's Constant (H) is an important parameter to describe environmental behaviour of atmospheric pollutants. Henry's Constant gives the ratio between vapour pressure (saturated) and the solubility of the compound in water.

Table 9: Ranges of Henry's constant for some organic environmental pollutants

Class of Compounds	Henry's Constant (atm·L/mol)
Halogenated C1 and C2 hydrocarbons	0.5-50
Alkylbenzenes (butylbenzene-benzene)	1-10
Chlorobenzenes (hexachlorobenzene-monochlorobenzene)	0.5-10
Phthalate esters	0.001-0.002
Polycyclic aromatic hydrocarbons (PAH)	0.005-1
Alipatic hydrocarbons (C18-C5)	10-10000
PCB	0.01-1

Henry's constant for PCB is in the range 0.8 atm L/mol for 2-monochlorobiphenyl and 0.018atm L/mol for decachlorobiphenyl and thus, in the same range like 2,3,7,8-Cl4DD. Henry's Law Constant is temperature dependent: H increases 10-fold in the temperature range 1450 °C. Within the same degree of chlorination, H increases with the umber of Cl atoms in ortho position [see Fiedler *et al.* 1994].

PCB Levels in the Environment

PCB have been identified in almost every environmental compartment or matrix. Detected levels depend on the nature and location of the particular environmental sample. However, congener-specific analytical procedures for qualitative and quantitative detection of PCB has not been developed as far as, for example, analysis of PCDD/PCDF.

When compared to other chemicals, PCB have very high KOW values: log KOW are in the range from 4.5 for monochlorobiphenyls to >8 for higher chlorinated PCB. Consequently, PCB tend to adsorb to unpolar surfaces and accumulate in lipophilic matrices along the aquatic and terrestrial food-chain. Some physical-chemical characteristics are given in Table 10.

Table 10: Influence of chlorine substituents on the chemical-physical properties of hydrocarbons
S = Water solubility, KOW = Octanol/water partition coefficient

Compound	Number of Chlorine Atoms	S (mg/L)	log KOW
Benzene	0	1,780	2.13
Hexachlorobenzene	6	0.006	6.18
Phenol	0	82,000	1.45
Pentachlorophenol	5	14	3.7
Biphenyl	0	5.9-7.5	3.89
PCB 209	10	0.000004	8.23
Dibenzo-*p*-dioxin	0	0.842	4.3
2,3,7,8-Cl4DD	4	0.000008	7
Cl8DD	8	0.0000004	8.2

Commercial PCB, as well as environmental extracts, contain complex mixtures of congeners. PCB mixtures found in environmental matrices usually do not resemble the commercial PCB mixtures. For example, a congener-specific analysis of PCB showed remarkable differences between a commercial Aroclor 1260 mixture and human breast milk [Safe 1990 and 1994, Norén and Lundén 1991]. This difference is due to the fact that the most abundant PCB in commercial mixtures are *ortho*-substituted congeners which are readily degradable. However, smaller amounts of the so-called „dioxin-like" PCB, namely the coplanar (= *non-ortho* substituted) and *mono-ortho* substituted congeners, are present in the commercial mixtures as well. The latter are very stable and resistant to biodegradation and metabolism. Moreover, it is well known that lower chlorinated PCB can volatilise and are, thus, more susceptible to atmospheric removal processes [Mackay *et al.* 1992].

Generally, the PCB levels found in environmental matrices are higher than the levels of PCDD/PCDF. This is due to the fact that besides thermal formation significant amounts of PCB were and still are released *via* diffuse emissions from industrial products.

Input of PCB into soil occurs - as for other lipophilic chemicals - either from spills, direct application of, *e.g.* sludges, or *via* dry and wet deposition. The organic carbon of soil is the natural sink for such unpolar lipophilic substances. Due to the strong affinity to organic carbon, PCB are quite immobile in soils. In combination with the persistence of the PCB, soils possess a memory effect and remember inputs long times ago as well as long-term diffuse inputs.

Henry's Law constants are 01 for mono- and dichlorobiphenyls, thus, these substances will be found preferentially in the gas phase and due to the low water solubility are not washed out with rainwater from the atmosphere. Higher chlorinated biphenyls are (completely) adsorbed to particulates and thus, can be removed from the atmosphere by capture of aerosols in rain drops. These two effects result in a relative accumulation of the lower chlorinated PCB in the atmosphere [Duinker and Bouchertall 1989]. Air concentrations are in the pg/m³ to ng/m³ range with lower levels in remote and rural areas. Background air levels in the USA were constant in the range of 1 ng/m³ over several years with tri- and tetrachlorinated congeners dominating.

Reports on the occurrence of PCB in fish, mussels, seals, sea birds and birds of prey first appeared in 1966, and in 1967, PCB were detected in human adipose tissue, albeit in low concentrations. In 1968, PCB from a leaking cooling system contaminated a rice oil tank at a food factory in Japan. As a result of the consumption of the contaminated rice oil which had reached the stores, 1,000 people fell ill with a disease subsequently known world-wide as Yusho Disease.

PCB in the Great Lakes display a more complex behaviour. They were found to volatilise where a river discharges relatively

high PCB loads into Green Bay, Wisconsin. Baker and Eisenreich [1990] calculated an average volatilisation rate of PCB from Lake Superior which approximately equals their atmospheric deposition. His findings support the conceptual model that these compounds permanently cycle between atmosphere and natural waters [Mackay *et al.* 1986]. According to this model, PCB dissolved in rain drops or sorbed to particulates are washed out of the atmosphere by rain. This input of PCB into surface waters results in a fugacity gradient towards the atmosphere, which in turn drives volatilisation.

Relatively new PCB data exist for sediments and suspended particles in German rivers. Along the river Saar it was found that close to locations with heavy industry (coal mining and steel industry) PCB and Ugilec (a commercial mixture of tetrachlorinated 2-methyl-diphenylmethanes) levels were higher than normal. Differentiation in depth showed that Ugilecs were only found in more recent sediments whereas PCB could be detected down to 1.2 m with higher concentrations in the older sediments [see Fiedler *et al.* 1994].

Environmental Fate of PCB

Biodegradation

Biodegradation by microorganisms may occur *via* three different mechanisms [for summary, see Fiedler *et al.* 1994]:
- Aerobic respiration in the presence of oxygen, involving reaction with mono- and dioxygenases; in a final step H_2O will be incorporated;
- Anaerobic respiration under exclusion of oxygen; inorganic substances such as nitrate, sulfate, and carbon monoxide act as electron acceptors;
- Fermentation under exclusion of oxygen; the organic compound to be degraded acts as electron acceptor.

Aerobic Degradation

In general, bacteria cannot use chlorinated aromatic hydrocarbons as substrate. Present knowledge assumes that bacteria growing on non-chlorinated biphenyl are capable to cause chemical reactions on the chlorinated ring system as well. However, some microorganisms are capable to use lower chlorinated PCB as C-source. Thus, *Acinetobacter sp. P6*, *Achromobacter sp.* B 218, and *Bacillus brevis* B 257 can grow on 4-chlorobiphenyl as the only carbon source. The main degradation product is 4-chlorobenzoic acid. In general, formation of chlorinated benzoic acids is the major degradation pathway for PCB. Further microorganisms capable for biodegradation of PCB belong to the class of *Acetobacter*, *Alcaligenes,* and *Pseudomonas*.

Some general conclusions can be drawn [Rochkind *et al.* 1986; for summary, see Fiedler *et al.* 1994]:
- Increasing number of chlorine substituents decreases biodegradation of PCB;
- Two chlorines in *ortho*-position at the same or on different aromatic rings of the biphenyl molecule significantly inhibit biodegradation (exception: 2,4,6-trichlorobiphenyl which is readily biodegradable by *Acinetobacter*);
- If there are chlorine substituents on both rings, the ring with less chlorine atoms will be hydroxylated first;
- Biphenyls that have chlorine substituents only on one ring system are metabolised more rapidly than a chlorobiphenyl with the same number of chlorine atoms but on both rings; thus, 3,4-dichlorobiphenyl will be better metabolised than 3´,4-dichlorobiphenyl;
- PCB metabolism is facilitated when a carbon atom with a chlorine substituent is between two unsubstituted carbon atoms;
- Higher chlorinated congeners having a 2,3,4-trichlorophenyl group, are resistant towards biological degradation;
- Ring cleavage occurs preferentially in the unsubstituted ring;
- PCB containing chlorines at positions 2 and 3, *e.g.* 2,2',3,3'-tetrachlorobiphenyl, 2,2',3,5'- tetrachlorobiphenyl, and 2,2',3',4,5-pentachlorobiphenyl are more readily biodegraded than other tetra- and pentachlorinated biphenyls.

Anaerobic Degradation

Polychlorinated biphenyls are extremely resistant to conventional aerobic transformation, but they will undergo anaerobic reductive dechlorination. Studies of PCB contamination in Hudson River sediment demonstrate that anaerobic environments yield markedly lower levels of tri-, tetra-, and pentachlorobiphenyls and higher levels of mono- and dichlorobiphenyls. Many of the less chlorinated PCB then are aerobically biodegradable because generally less toxic than highly chlorinated PCB.

For PCB, the degradation rate is inversely related to the degree of chlorination; thus, highly chlorinated congeners are more readily dechlorinated than lower chlorinated congeners.

There is evidence that not only the number of chlorine substituents determines degradation rates, but also their position. Reductive dechlorination predominantly reduces chlorine in *meta-* and *para-*positions, resulting in accumulation of the *ortho-*chlorinated congeners. Addition of organic substrates, such as methanol, glucose or acetone stimulated dechlorination, whereas little stimulation was observed in cases with no organic additives [see Fiedler *et al.* 1994].

Metabolism

PCB do not have reactive functional groups; thus, these lipophilic molecules have to be hydroxylated first to make them more polar and consequently subject for excretion. The rate limiting step in the elimination of PCB is that of metabolism, which primarily occurs by the hepatic P-450-dependent monoxygenase system. Hydroxylated products are the major PCB metabolites and, based on available studies, it can be concluded that hydroxylation mainly occurs at *para* or *meta* positions if these sites are unsubstituted. The chlorine content, the substitution pattern and the presence of certain isoenzymes of the cytochrom-P-450 system are important factors to determine the transformation rate of PCB [see Fiedler *et al.* 1994]. In general, metabolisation of PCB decreases with increasing number of chlorine atoms present and with decreasing number of adjacent unsubstituted carbon atoms. Isoenzymes capable to metabolise Phenobarbital (PB) were found to metabolise the not dioxin-like PCB whereas coplanar, non-*ortho* or *mono-ortho*-substituted (dioxin-like) PCB can induce isoenzymes capable to metabolise 3-methylcholanthrene (MC) [Safe 1994]. Commercial PCB, such as Aroclor 1254, induce both, MC and PB-inducible monooxygenases. Besides hydroxylation and subsequent conjugation, sulfur-containing metabolites, *e.g.* methyl sulfones, and partially dechlorinated metabolites have also been identified. Methyl sulfones have been shown to selectively accumulate in the Clara cells of rat lung and in lung tissues of mice. Methyl sulfonyl metabolites of 2,4´,5-, 2,2´,4,5-´, and 2,2´,4,5,5-PCB have also been found in liver, adipose and fetal tissues and have been identified in environmental samples and in human milk [Ahlborg *et al.* 1992].

Due to the low transformation and excretion rates of PCB, certain congeners accumulate in organisms. Persistent congeners, such as 2,2',4,4',5,5'-hexachlorobiphenyl (PCB 153) were found to promote tumours in rats, whereas 2,2',3,3',6,6'-hexachlorobiphenyl (PCB 136) is easily degraded.

Bioconcentration

Substances of low biological degradability tend to accumulate throughout trophic levels of the food net. For example, concentrations of total PCB increases with the trophic level. Only concentrations of PCB in sediments are higher than levels in the subsequent trophic levels. It has been demonstrated that chlorinated dibenzo-*p*-dioxin and dibenzofuran congeners accumulate with considerable species differentiation. Contribution of the dioxin-like PCB congeners # 77, 105, and 126 to the total TEQ is substantially greater than that of PCDD/PCDF (even in cases of known PCDD/PCDF contamination). In all cases an exchange between trophic levels (sediment algae plankton planktivores piscivorous fish piscivorous birds) resulted in an increase of both, total PCB concentration and dioxin-like TEQ (for TEF/TEQ of PCB, see section 5) [Fiedler *et al.* 1994].

Toxicity of PCB

Commercial mixtures, as well as the individual PCB congeners, elicit a broad spectrum of biochemical and toxic responses, some of which are similar to those caused by 2,3,7,8-tetrachlorodibenzo-*p*-dioxin. Due to the fact that dioxin-like compounds normally exist in environmental and biological samples as complex mixtures of congeners, the concept of toxic equivalents (TEQ) has been developed to simplify risk assessment and regulatory control. In applying this concept, relative toxicities of dioxin-like compounds in relation to the reference compound (2,3,7,8-tetrachlorodibenzo-*p*-dioxin, 2,3,7,8-Cl4DD) were determined on the basis of results obtained in *in vivo* and *in vitro* studies (Table 11). Three coplanar PCB, namely 3,3',4,4'-tetrachlorobiphenyl, 3,3',4,4',5-pentachlorobiphenyl, and 3,3',4,4',5,5'-hexachlorobiphenyl, exhibit dioxin-like effects, such as *Ah*-receptor agonist activity [Safe 1990 and 1994, Ahlborg *et al.* 1992 and 1994]. PCB 170 and PCB 180 are included because they are active as inducers of EROD activity and are present in significant amounts in environmental samples.

To illustrate the consequences of the recommended PCB-TEF, the contribution of dioxin-like PCB and PCDD/PCDF to the total TEQ was calculated for some matrices (Table 12). As can be seen from Table 12, the contribution from the PCB to the total TEQ for PCDD/PCDF plus PCB is between 50% and 200%.

Table 11: Proposed TEF for coplanar and *mono-* and *di-ortho*-substituted PCB [Ahlborg *et al.* 1994]

Congener	Substitution	IUPAC No.	TEF
Non-ortho substituted PCB:	3,3',4,4'-TetraCB	77	0.0005
	3,3',4,4',5-PentaCB	126	0.1
	3,3',4,4',5,5'-HexaCB	169	0.01
Mono-ortho substituted PCB:	2,3,3',4,4'-PentaCB	105	0.0001
	2,3,4,4',5-PentaCB	114	0.0005
	2,3',4,4',5-PentaCB	118	0.0001
	2',3,4,4',5-PentaCB	123	0.0001
	2,3,3',4,4',5-HexaCB	156	0.0005
	2,3,3',4,4',5'-HexaCB	157	0.0005
	2,3',4,4',5,5'-HexaCB	167	0.00001
	2,3,3',4,4',5,5'-HeptaCB	189	0.0001
Di-ortho substituted PCB	2,2',3,3',4,4',5-HeptaCB	170	0.0001
	2,2',3,4,4',5,5'-HeptaCB	180	0.00001

Table 12. Toxic equivalents (TEQ) calculated for fish, cow milk, and human milk samples using the interim WHO/ICPS TEF [Ahlborg *et al.* 1994]

TEQ	Mother's Milk	Cow Milk	Salmon

Sum of TEQ for *non-ortho* PCB	10.3	2.4	67.7
Sum of TEQ for *mono-ortho* PCB	10.1	0.4	46.8
Sum of TEQ for *di-ortho* PCB	0.6	0.04	8.3
Total TEQ for PCB	21.0	2.8	122.8
Total TEQ for PCDD/PCDF	20.6	5.6	56.0

Global Distribution - Long-Range Transport

Today it is known that many chlorinated organics and other stable compounds are distributed at a global scale through atmospheric transport. A general tendency in these transport patterns is that different substances are evaporated and spread to the atmosphere at latitudes with warmer climates and then condense and fall-out closer to the poles (= global condensation). Consequently, areas close to the North and the South pole receive a disproportionate share of this fall-out. An indication of this phenomenon is that several chlorinated pesticides, long banned in countries, such as Sweden, are found - although at relatively low levels - in environmental compartments of this country. Examples of these substances are chlordane, toxaphene, and hexachlorocyclohexane; thus, compounds that are still being produced in other countries. For PCB, an annual fall-out of 4 tons was estimated for Sweden in 1994.

The presence of persistent polychlorinated compounds in remote areas such as the Arctic and the Antarctic has been reported. PCB, PCDD/PCDF, HCH, and HCB were found in marine organisms such as seal blubber and pinniped milk as well as in lake and sea sediments. The occurrence of mainly man-made organochlorines in regions far away from industrialised and densely populated areas indicates that atmospheric transport is an important route to disperse these compounds. All three groups of compounds, PCB, PCDD/PCDF, and HCB, have the same source areas: Densely populated and industrialised regions. From these source regions, the organochlorines are transported *via* various mechanisms. Norstrom (Environment Canada) summarised the present knowledge on the occurrence of persistent chlorinated organic compounds in the Arctic aquatic environment as follows [see Fiedler *et al.* 1994]:

Chlorinated hydrocarbons (CHC), such as the pesticides chlordane and toxaphene (polychlorinated camphenes, PCC), hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB) and DDT, and industrial chemicals, such as polychlorinated biphenyls (PCB) have been identified in air, snow, ocean water, and biota in the marine ecosystem. Although most open uses of these chemicals were curtailed in many industrial countries, a considerable fraction of these compounds is still cycling in the ecosphere. Thus, it was estimated that 20 % of the world production of PCB, 230,000 tons, are present in the upper layers of the ocean, and 790 tons were in the open ocean atmosphere. Large quantities of chlorinated pesticides continue to be used in less developed countries, especially in the Southern hemisphere. Although there is little information on the production amounts and releases of organochlorines from Russia and China, these areas are undoubtedly major contributors to the environmental burden with CHC.

Chlorinated pesticides and PCB have sufficiently high vapour pressures that they readily volatilise when spread over a large surface area such as soil or water. Atmospheric residence time of PCB has been calculated to be in the order of a few months. Henry's Law constants of the above mentioned compounds are in the range of 0.1-50 Pa·m³/mol and thus, will allow that these substances will evaporise and cycle back and forth between land or surface waters and air. These processes lead to a global distribution. The „cold finger effect" will result in the fact that the Arctic and the Antarctic regions will become the sinks for organochlorines due to the distillation of these compounds from warmer to colder regions.

Average concentrations of the major classes of CHC in the marine environment of the Arctic are given in Table 13. Hexachlorocyclohexanes (HCHs) and hexachlorobenzene (HCB) are the dominant CHC in air, followed by the PCC. DDTs, chlordane, and PCB are one order of magnitude lower. Because of its higher Henry's Law constant, HCB is less dominating in snow and seawater. The high levels of HCH in the Arctic Sea water support the distillation theory. Moreover, sources in Asia

might have a contribution. The effect of the higher lipophilicity of compounds such as DDTs, chlordanes, and PCB goes along with bioconcentration. The concentrations of these compounds increase along the aquatic food-chain from plankton to beluga whales. The only exception are the HCHs which do not tend to strongly bioconcentrate (low KOW).

Generally, the concentrations in the Antarctic are lower than the levels in the Arctic. This finding is reasonable as more than 80% of the industrialised regions which act as sources for PCDD/PCDF and PCB are located in the Northern hemisphere. The slow inter-hemispheric air exchange (about 1-2 years) reduces the atmospheric transport of semi-volatile compounds from the Northern to the Southern hemisphere. Oehme and co-workers analysed air samples from both regions and found some interesting differences: Levels of hexachlorocyclohexanes (HCH) and hexachlorobenzene (HCB) were lower in the Antarctic air. Whereas in the Arctic air, -HCH was dominating, the -isomer dominated in the Antarctic. This indicates that preferentially the pure -HCH has been applied in the Southern hemisphere whereas in the northern hemisphere more of the technical mixture (80-85% -HCH) was used. HCB is mostly of anthropogenic origin - from incomplete combustion processes and utilisation as pesticide - and its atmospheric half-life time is 1-2 years leads to an almost homogeneous distribution on both hemispheres. Four chlordane compounds were identified in similar concentrations at both hemispheres. The PCDD/PCDF pattern found in Antarctic fur seal blubber was significantly different from that found in Arctic ringed seals and harp seals. Reasons might be differences in the emission patterns in the Northern and the Southern hemispheres, different food habits or inter-species variations. Amongst the PCB pattern, PCB 77, PCB 126 and PCB 169 were similar in Antarctic fur seals when compared to the Arctic harp seal. However, the levels - in TEQ - were lower in the Antarctic seals by a factor of about 5. This is less than the 12 orders of magnitude reported in earlier studies [see Fiedler *et al.* 1994].

Table 13: Average concentrations of organochlorines in the Arctic marine environment
NA = Not available

Matrix, Dimension	HCHs	HCB	PCC	DDT	Chlordanes	PCB
Air, ng/m³	0.58	0.19	0.044	<0.001	0.006	0.014
Snow, ng/L	1.72	<0.002	0.085	<0.01	0.06	0.086
Seawater (surf.), ng/L	4.3	0.028	0.36	<0.001	0.004	0.007
Seawater (deep), ng/L	0.51	0.01	0.11	<0.002	0.005	<0.014
Zooplankton, µg/g lipid	0.08	0.02	0.06	0.06	0.06	0.11
Amphipods, µg/g lipid	0.5	0.17	NA	<0.35	0.43	<0.44
Cod, µg/g lipid	0.58	0.2	1.84	0.26	0.19	0.23
Beluga, µg/g lipid	0.25	0.5	3.11	2.82	1.76	3.79
Ringed seal, µg/g lipid	0.23	0.03	0.32	0.5	0.4	0.55
Polar bear, µg/g lipid	0.51	0.27	ca. 0.4	0.4	3.7	5.4
Human milk, µg/g lipid	NA	0.14	NA	1.21	NA	1.05

Last but not least, it should be mentioned that distribution of chlorinated organic compounds also occurs *via* economic pathways and international trade of goods. Examples are textiles, leather, wood, packaging materials, *etc.* Consequently, ban of a given chloroorganic compound by one or a few countries - as, *e.g.* for pentachlorophenol in Germany, DDT, PCB in several

industrialised countries - does not prevent a country from „pollution" with the given compound or its contaminants (*e.g.* PCDD/PCDF). Moreover, it is difficult to control the concentrations of chemicals in these goods that pass the country's borders.

The same thoughts have to be applied for transboundary atmospheric „imports" of unwanted pollutants: Stringent emissions limits set by own authorities do not prevent the country from importing atmospheric contaminants from the neighbour. Thus, in many aspects, the spread of persistent chloroorganic chemicals ask for international solutions.

PCB Exposure: Occupational and General

Occupational

No studies have been conducted on the emissions during the production of PCB. It is assumed, however, that they were low. Major quantities reached the environment in the past through, for example, improper disposal of PCB-containing products and transformer fires. Other emission sources are landfills, small PCB-containing capacitors in household appliances and PCB-containing sealants for buildings. Inputs of PCB to the environment can also occur through application of sewage sludge and sediments of water bodies. Products from the reprocessing of waste oil are another source from which inputs occurred.

When the regulations for handling and transport of PCB and PCB-containing waste are strictly enforced (as required in the German regulation = TRGS), there is little or no increased occupational exposure to be expected.

Exposure of the General Population

In Germany mother's milk has been analysed on a large scale (about a thousand analyses per year) for various toxic substances like hexachlorobenzene, DDT, and PCB. Due to the continuous change in analytical procedures for PCB-determination it is difficult to compare values obtained for example for mother's milk before 1984 with those of recent years. Regardless of this analytical problem it is evident that since about 1980 the maximum values for PCB concentrations have been decreasing constantly. The range of PCB-concentrations in mother's milk in the last ten years is between 0.5 and 2.5 mg/kg milk fat. The average concentrations have been decreasing constantly since 1984. This is shown in Figure 2 [NRW 1996]. However, as can be seen from Figure 3, there was a major decrease in the mother's milk concentrations in the mid and late 1970s whereas in the 1980s the concentrations remained on the same level. More recently, the concentrations seem to further decrease.

Figure 2: PCB in mother's milk - Mean concentrations [NRW 1996]

Figure 3: Time trend of PCB concentrations in mother's milk [Brune and Fiedler 1996]

From the possible transfer paths of human exposure through direct skin or air contact with technical PCB mixtures, meat and meat products, milk and milk products, fish and fish products, ambient and indoor air, the direct uptake of technical PCB can be considered as negligible. In Germany, indirect exposure pathways *via* the terrestrial food-chain dominate the human intake. PCB emitted into the air from combustion sources or re-entrained from reservoir sources (contaminated land, vegetation, surfaces in general) deposit on grass and other fodder plants which are eaten by cattle and thus, contaminating milk (and products) and meat (and products). Due to the many PCB contaminations of sediments from the years before 1980, PCB can still be found in fish. The terrestrial and the aquatic food-chains are the most important routes for human exposure to PCB.

The question arises to what extent there exist primary sources of PCB input into the environment by *de novo* synthesis of PCB by thermal sources, analogous to dioxin synthesis. At least for Germany we estimate that the PCB uptake *via* meat and milk and respective products originates to a major extent by the latter source [Hagenmaier 1996].

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