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Towards a global historical emission inventory for selected PCB congeners — a mass balance approach 1. Global production and consumption

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Abstract

Information on the historical global production and consumption of polychlorinated biphenyls (PCBs) is urgently needed for estimating PCB fluxes to the environment and for interpreting global contamination patterns by these pollutants. This study presents the methodology, principal uncertainties and selected results from an inventory, aiming to quantify the global production and consumption of total PCBs as well as 22 PCB congeners. The available data on the historical production of PCBs and the chemical composition of various technical mixtures have been compiled from the literature. For some producers with less detailed information, the production of individual PCB constituents has been estimated to derive a global estimate for individual homologues and selected congeners. Information on imports, exports and consumption, as well as restrictions on production and imports, has further been compiled for individual countries. These data, along with assumptions on the trade between countries and regions, have been utilised to derive an estimate of the global historical consumption pattern. Although there are substantial uncertainties involved in these estimates, important aspects governing the large scale temporal and spatial patterns are most likely captured in these estimates. In particular, the information on imports and exports for the principal users of PCBs around the time of peak production is considered to be fairly reliable. The estimates account for a reported historical global production of ~1.3 million t PCBs, more than 70% of which are tri-, tetra- and pentachlorinated biphenyls. The results further suggest that almost 97% of the global historical use of PCBs have occurred in the Northern Hemisphere.

Keywords: POPs; PCBs; Homologues; Congeners; Sources; Production; Consumption; Global; Historical

1. Introduction

Environmental contamination by polychlorinated biphenyls (PCBs) was recognised more than 30 years ago when Sören Jensen detected PCBs in pike from Sweden (Jensen, 1966). Since then, numerous studies have detected PCBs in various compartments of the environment (e.g. Edwards, 1971; Kalmaz and Kalmaz, 1979; Waid, 1986) and the occurrence of PCBs in remote areas, such

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as the Arctic (e.g. AMAP, 1998) is evidence for the long-range atmospheric transport of these contaminants (Oehme and Manø, 1984; Oehme, 1991; Harner et al., 1998). Today, PCBs are considered an environmental problem of global proportions.

Even though the production of these contaminants has stopped, PCBs continue to be detected in environmental samples from around the world (e.g. Iwata et al., 1994; AMAP, 1998). Many studies have sought to understand historical PCB contamination trends through the analysis of dated sediment (e.g. Christensen and Lo, 1986; Sanders et al., 1992; Bruckmeier et al., 1997) and peat cores (Rapaport and Eisenreich, 1988). Several of these studies have indicated that the trends in environmental concentrations have followed the trends in production and use of PCBs. Historical data on the global production and usage of PCBs are, thus, urgently needed for the interpretation of historical, present and future contamination levels around the world (Cummins, 1988; Tanabe, 1988; Voldner and Li, 1995; Wania and Mackay, 1996; Vallack et al., 1998). Furthermore, quantitative knowledge of the global historical consumption is a prerequisite for estimating atmospheric emissions and eventually establishing source–receptor relationships for intentionally produced PCBs on a global scale. Due to differences in lifetime and fate of individual PCBs in the environment, estimates of the historical production of PCBs have to be done on a congener specific basis. The overall aim of this study was to present a quantitative estimate of the historical consumption of selected PCB congeners. Specifically, we set out to:

1. estimate the historical global production of selected PCB congeners (*temporal pattern*);
2. estimate the historical global pattern of consumption (*spatial pattern*); and
3. provide input for a global PCB emission model, presented in an accompanying paper (Breivik et al., 2002).

2. Methods

The general molecular formula for the PCBs is $C_{12}H_{10-n}Cl_n$, where n could be any number from 1 to 10. There are, thus, 10 different PCB homologues, dependent on the number of chlorines and 209 different PCB congeners, dependent on the position of the chlorines on the molecule. Not all congeners have been identified in commercial products or technical mixtures. The numbering system proposed by Ballschmiter and Zell (1980) has been adapted by the International Union of Pure and Applied Chemists (IUPAC), and is frequently used to refer to various congeners. In this system, individual PCB congeners are assigned a number, ranging from PCB-1 (2-CB) to PCB-209 (2,2′,3,3′,4,4′,5,5′,6,6′-CB). This numbering system is also used here, although with minor revisions (Hillery et al. 1997). In this work, 22 individual PCB congeners were studied. These are the same congeners as selected in the EU GlobalSOC project (ENV4-CT97-0638), or more specifically PCB-5 (2,3-DiCB), PCB-8 (2,4′-DiCB), PCB-18 (2,2′,5-TriCB), PCB-28 (2,4,4′-TriCB), PCB-31 (2,4′,5-TriCB), PCB-52 (2,2′,5,5′-TetCB), PCB-70 (2,3′,4′,5-TetCB), PCB-90 (2,2′,3,4′,5-PenCB), PCB-101 (2,2′,4,5,5′-PenCB), PCB-105 (2,3,3′,4,4′-PenCB), PCB-110 (2,3,3′,4′,6-PenCB), PCB-118 (2,3′,4,4′,5-PenCB), PCB-123 (2′,3,4,4′,5-PenCB, PCB-132 (2,2′,3,3′,4,6-HexCB), PCB-138 (2,2′,3,4,4′,5′-HexCB), PCB-149 (2,2′,3,4′,5′,6-HexCB), PCB-153 (2,2′,4,4′,5,5′-HexCB), PCB-158 (2,3,3′,4,4′,6-HexCB), PCB-160 (2,3,3′,4,5,6-HexCB), PCB-180 (2,2′,3,4,4′,5,5′-HepCB), PCB-194 (2,2′,3,3′,4,4′,5,5′-OctaCB) and PCB-199 (2,2′,3,3′,4,5,5′,6′-OctaCB).

We proceeded by first collecting from the literature data on the production of total PCBs as well as of various technical PCB mixtures. Secondly, these data were combined with data on the composition of these technical mixtures to estimate the production of individual homologues and congeners. To fill gaps in the data, assumptions had to be made sometimes concerning the homologue and congener composition (Section 3.1). The global consumption pattern was assessed by compiling information on imports, exports and consumption of PCBs for individual countries and years. Reliable information is available only for countries with a historically high consumption of PCBs. For

Table 1
Total PCB production in t as reported in the literature

Producer	Country	Start	Stop	Amount	Reference
Monsanto	USA	1930	1977	641 246	de Voogt and Brinkman (1989)
Geneva Ind.	USA	1971	1973	454	de Voogt and Brinkman (1989)
Kanegafuchi	Japan	1954	1972	56 326	Tatsukawa (1976)
Mitsubishi	Japan	1969	1972	2461	Tatsukawa (1976)
Bayer AG	West Germany	1930	1983	159 062	de Voogt and Brinkman (1989)
Prodelec	France	1930	1984	134 654	de Voogt and Brinkman (1989)
S.A. Cros	Spain	1955	1984	29 012	de Voogt and Brinkman (1989)
Monsanto	U.K.	1954	1977	66 542	de Voogt and Brinkman (1989)
Caffaro	Italy	1958	1983	31 092	de Voogt and Brinkman (1989)
Chemko	Czechoslovakia	1959	1984	21 482	Schlosserová (1994)
Orgsteklo	USSR (Russia)	1939	1990	141 800	AMAP (2000)
Orgsintez	USSR (Russia)	1972	1993	32 000	AMAP (2000)
Xi'an	China	1960	1979	8000	Jiang et al. (1997)
Total		1930	1993	1 324 131	

other countries, assumptions had to be made on the trade between various countries and regions, using the Gross Domestic Product as a surrogate parameter (Section 3.2).

3. Results and discussion

3.1. Global production

3.1.1. PCB production process

The production of PCBs involves the chlorination of biphenyl in the presence of a catalyst. Depending on the reaction conditions, the degree of chlorination varies between 21% and 68% chlorine on a weight-by-weight basis (e.g. Ahlborg et al., 1992). The homologue profile for most technical formulations shows a normal distribution around the mean chlorine content (e.g. Takasuga et al., 1996, see also Table 2). This implies that these mixtures generally contain only a certain 'range' of PCB homologues and congeners as indicated by the chlorine content. However, some other technical formulations, such as Aroclor 1232, do not show this typical distribution, suggesting that they consist of more than one technical mixture (Frame, 1997).

3.1.2. Total global PCB production

A review of the literature data was undertaken to obtain the most reliable production figures for the major producers of PCB in various countries. A previous study had estimated the cumulative global production to be on the order of 1.5 million t (de Voogt and Brinkman, 1989). Most of the information presented in Table 1 is adapted from this compilation of data. The figures shown in Table 1 add up to a reported total global production of 1.324 million t between 1930 and 1993. Most likely the true cumulative global production has been higher, as there were factories in Poland (Falandysz et al., 1992), Eastern Germany (de Voogt and Brinkman, 1989) and Austria (Fiedler, 1997) that produced PCBs in unknown amounts. Although the data presented here might be lower than the real figure, it seems likely that most of the global historical production is accounted for in these estimates. For most producers within the OECD countries, data on the total amounts produced are generally reported for 5-year periods from 1955 to 1984. In addition, annual production data are available for the same countries from 1973 to 1980 (de Voogt and Brinkman, 1989). For producers outside the OECD, there is only limited information on the annual production from various plants. Production data reported for a period in excess of one year (e.g. a 5-year period) were uniformly distributed over that period (*temporally flat distributed*, see also Fig. 3a). These data show that Monsanto (USA) has been responsible for

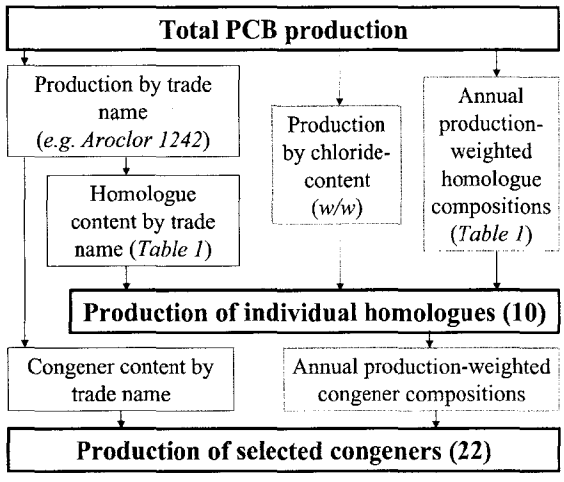

Fig. 1. Overview of the relationship between the annual production of total PCBs and the production of individual homologues and congeners from various producers. Details of the approach are discussed in Section 3.1.3.

almost 50% of the known reported historical production of PCBs. Bayer (West Germany), Prodelec (France) and Orgsteklo (Russia) have each contributed individually with more than 10% of the historical production.

3.1.3. Estimated production of individual homologues and congeners

The overall methodology for estimating the production of individual PCB homologues and congeners is depicted in Fig. 1. Whenever possible, information on the production of individual technical mixtures (e.g. Aroclor 1242), and their chemical composition was used to determine the homologue and congener production over time. However, as this information was not available for many producers, the annual production of individual homologues and congeners could only be estimated in this manner for Monsanto (USA) and Bayer (West Germany) (see below and Fig. 1). For other cases, we estimated a set of annual production-weighted default compositions (i.e. a fraction between 0 and 1). These default compositions or fractions take into account the temporal shift in homologue and congener production that occurred during the period of peak production (i.e. they were varying from 1955 to 1983).

The homologue composition (by wt.%) of various technical mixtures is given in Table 2, along with the estimated maximum and minimum default homologue composition. The annual production-weighted congener default compositions were estimated in a similar manner based on data of the technical mixtures produced by Monsanto (USA) and Bayer (West Germany) (data not shown).

As the availability of data for the different producers varied immensely, an individual approach was necessary to maximise the utilisation of available information and to minimise the use of uncertain assumptions. For the sake of transparency, details of the approach adopted for each individual producers are given in the following paragraphs. Although this might appear unnecessary, we hope that such detailed documentation will eventually facilitate future improvements to these estimates.

3.1.3.1. USA. The data compiled by de Voogt and Brinkman (1989) account for a total production of 641 699 t of PCBs in the USA. Monsanto was by far the most important producer, while the only other PCB-producing company in the USA (Geneva Industries) has a reported production of only 454 t.

The two last numbers in the names of the technical Aroclor mixtures by Monsanto refer to the weight percentage of chlorine (Table 2). For example, Aroclor 1260 should contain approximately 60 wt.% chlorine. Two notable exceptions to this numbering system are Aroclor 1016, which is a technical mixture derived by distillation of Aroclor 1242, and Arcoclor 1232 which is a blend of approximately equal proportions of Aroclors 1221 and 1242 (Frame, 1997). The congeneric composition of Aroclors 1016, 1221, 1232, 1242, 1254, 1260 and 1262 were based on data compiled by Frame (1997), who reported the results of a collaborative study in which the composition of six Aroclor mixtures (1016, 1221, 1242, 1254, 1260 and 1262) was determined using 18 gas chromatographic systems. Nine systems employed ECD detection, and the remaining half-used MS-

Table 2
Content of the 10 homologues of PCBs in various technical mixtures as used in the calculations (in wt.%)

Technical mixture		Mono-	Di-	Tri-	Tetra-	Penta-	Hexa-
Aroclor 1221		43.8	27.9	4.4	2.6	0.5	0.2
Aroclor 1232		26.5	23.9	27.0	18.7	3.5	0.3
Aroclor 1016		0.7	17.1	53.6	27.7	0.8	0.1
Aroclor 1242		0.3	14.7	42.1	33.9	8.1	0.8
Aroclor 1248		0.2	2.5	22.8	51.7	20.5	2.0
Aroclor 1254		–	0.5	0.7	18.3	55.6	22.0
Aroclor 1260		–	0.1	0.3	0.9	9.9	43.5
Aroclor 1262		–	0.2	1.2	1.1	3.9	28.1
Clophen A30		–	19.6	48.1	25.0	6.2	1.2
Clophen A40		–	0.2	17.3	50.5	25.8	5.0
Clophen A50		–	–	0.2	17.6	51.2	26.8
Clophen A60		–	–	–	0.7	16.2	49.1
Sovol		–	–	1.0	23.0	53.0	22.0
TCB		–	14.0	49.0	32.0	4.0	1.0
Delor 123		9.0	63.0	26.0	2.0	–	–
Delor 103		1.0	10.0	60.0	26.0	3.0	–
Default compositions	Min	0	6.6	19.5	22.9	6.2	1.2
	Max	1.0	19.6	48.1	28.4	22.5	17.0

Technical mixture		Hepta-	Octa-	Nona-	Deca-	Notes
Aroclor 1221		-	–	–	–	A
Aroclor 1232		0.1	–	–	–	A
Aroclor 1016		–	–	–	–	A,B
Aroclor 1242		0.1	–	–	–	A,B
Aroclor 1248		0.3	0.1	–	–	A
Aroclor 1254		2.5	0.4	–	–	A,B
Aroclor 1260		36.1	8.3	0.9	–	A,B
Aroclor 1262		45.3	18.5	1.7	–	A
Clophen A30		–	–	–	–	B
Clophen A40		1.2	–	–	–	B
Clophen A50		3.6	0.6	–	–	B
Clophen A60		27.8	5.8	0.5	–	B
Sovol		1.0	–	–	–	C
TCB		–	–	–	–	C
Delor 123		–	–	–	–	D
Delor 103		–	–	–	–	D
Default compositions	Min	0	0	0	0	E
	Max	9.3	2.1	0.2	0	E

The original data from the literature were scaled to yield 100%, except Aroclor 1221 (see text). [A] Frame (1997); [B] Schulz et al. (1989); [C] Ivanov and Sandell (1992); [D] de Voogt and Brinkman (1989); [E] Annual production-weighted compositions (or ratios) are based on the estimated production from Bayer AG (West Germany) and Monsanto (USA). Only the minimum and maximum compositions are shown.

SIM or full-scan MS ion-trap measurements (see Frame, 1997 for details). Average weight percentage of individual congeners in each of these Aroclors was given for both ECD and MS systems. Frame (1997) also reported data on the composition of Aroclor 1232 and 1248, analysed by one of the participating laboratories in his study (referred to as JWC). In addition, data for Aroclor 1016, 1242, 1254 and 1260 from a previous study (Schulz et al. 1989) were included.

We used a weighted average of these data to calculate the congeneric composition of the Aro-

clor production. These compositions, reported in Table 2, are thus based on 19 different measurements for Aroclor 1016, 1242, 1254 and 1260, 18 different determinations for 1221 and 1262, and a single determination for 1232 and 1248. These data were also used to estimate the homologue composition of each individual mixture and scaled to yield a total of 100%. The composition of the lighter chlorinated Aroclor 1221 was not scaled this way, because it presumably contains significant amounts of biphenyl due to incomplete chlorination.

The amounts of various Aroclors (1016:1242:1248:1254:1260 and 'Other Aroclors') sold in the USA from 1957 to 1975 are available from de Voogt and Brinkman (1989). These data were used to estimate and scale the production of individual Aroclors, assuming that the production of various Aroclors was equal to the annual fraction of sold amounts. For the years prior to 1957 and after 1975, we used the estimated fractions for 1957 and 1975, respectively. The sold amounts of the 'Other Aroclors' (1221 + 1232 + 1262 + 1268) was generally below 3% during the investigated time-period (de Voogt and Brinkman, 1989). In the absence of information on the chemical composition of Aroclor 1268, we assumed that 'Other Aroclors' was a mixture of 1221, 1232 and 1262 (1:1:2). As the 'Other Aroclors' include the heavily chlorinated mixture 1268 in unknown relative amounts, it is likely that we are underestimating the produced amounts of some of the more chlorinated PCB homologues and congeners. Monsanto also produced a technical mixture called Aroclor 1270 (de Voogt and Brinkman, 1989). Due to the lack of data on both production and chemical composition, it could not be included in this estimate.

For the production of PCBs by Geneva Industries we relied on the default homologue and congener composition (see Fig. 1).

3.1.3.2. West Germany. PCBs were produced in West Germany by Bayer AG as Clophens (A30, A40, A50, A60). The data presented by de Voogt and Brinkman (1989) account for a historical production of 159,062 t PCB. According to this reference, the approximate wt.% of chlorine in various trade mixtures were: A30 (40–42%), A40 (48%), A50 (52–54%) and A60 (60%). Fiedler (1997) presented production data by degree of chlorination (39, 42.5, 47, 48.5, 54, 55 and 60% Cl (w/w)) for the period from 1974 to 1983. As the amounts of individual Clophens produced were not available to us, we assigned the data presented by Fiedler (1997) to the corresponding Clophen-mixture, based on the degree of chlorination in order to estimate fractions of the technical mixtures produced annually. For data prior to 1974, we assumed that the various mixtures were produced in the same relative quantities as in 1974. The homologue and congener production could then be estimated based on the homologue and congener content of A30, A40, A50 and A60 reported by Schulz et al. (1989).

3.1.3.3. Japan. Approximately 96% of the total Japanese PCB production was by Kanegafuchi Chemical Co. Ltd (Tatsukawa, 1976), which produced a series of PCB mixtures called Kanechlors (KC). According to Tatsukawa (1976), the chlorine content of KC-300, KC-400, KC-500 and KC-600 corresponds to the Aroclors 1242, 1248, 1254 and 1260, but the homologue composition is different. Although the chemical composition of Kanechlors has been at least partly determined (Kannan et al. 1992; Takasuga et al., 1996), this information could not be used because the individual amounts of Kanechlors produced are unknown to us. Therefore, we used annual production data for various homologues reported by Tatsukawa (1976) for the years 1961 to 1971 as tri-CBs and lower, tetra-CBs, penta-CBs and hexa-CBs and higher. For the two clustered homologue groups (tri-CB and lower, hexa-CB and higher), we assumed that the internal homologue production was determined by the percentage of possible congeners. For example, the group of tri-CB and lower includes 39 possible congeners, while there are three possible congeners within the group of mono-CB. Thus, 7.7% of the clustered tri-CBs and lower was assumed to be mono-CB, etc.

The production of individual congeners was estimated using the congeneric default composition (Fig. 1). These percentages were multiplied by the reported or estimated annual production of individ-

ual homologues as described above. This procedure clearly introduces high uncertainties to the estimated annual production of individual congeners, but it seems reasonable to assume that this approach avoids emphasis on congeners that are rarely formed during the production process. For the other producer in Japan (Mitsubishi), we applied default homologue and congener compositions to total PCB production figures reported by Tatsukawa (1976).

3.1.3.4. Czechoslovakia. Schlosserová (1994) reported that 14 140 t of PCB was produced in Czechoslovakia as Delor 103. In addition, 4381 t of Delor 106 (similar to Aroclor 1260) and 2961 t of other PCB mixtures were produced from 1959 to 1984. de Voogt and Brinkman (1989) reported that approximately 6000 t were produced annually before 1968, suggesting higher production figures than those reported by Schlosserová (1994). However, we have chosen to use the more detailed and recent information provided by Schlosserová (1994), assuming a uniform annual production of 826 t total PCBs throughout the period.

To estimate the homologue production, we used the homologue composition of Delor 103 as reported by de Voogt and Brinkman (1989) and assumed that Delor 106 has the same composition as Aroclor 1260 (Table 2). For the remaining 4381 t, we assumed that its composition is that of Delor 123, a very light PCB formulation described by de Voogt and Brinkman (1989). We further used the congeneric default composition for estimating the production of individual congeners.

3.1.3.5. United Kingdom. In the UK, PCBs were manufactured under the trade name Pyroclor (de Voogt and Brinkman, 1989). As we did not have reliable information on the relative production volume, or chemical composition of the Pyroclors, we applied the default homologue and congener compositions. The UK factory was owned by Monsanto Industrial Chemicals Co., which also was the major producer of PCBs in USA. Similarities with the production process at Monsanto in the US are thus likely.

3.1.3.6. Soviet Union. Estimates of the historical production of PCBs in the former Soviet Union are available from Ivanov and Sandell (1992) and more recently from AMAP (2000). According to this latter source, PCBs were produced under three different brand names (Sovol, Sovtol and TCB) at two different factories in the vicinity of Moscow (Orgsteklo and Orgsintez).

Sovol is reported to have a chemical composition fairly close to that of Aroclor 1254 (Ivanov and Sandell, 1992; Takasuga et al. 1996). However, another study suggests that it only resembles Aroclor 1254 to a limited extent (Kannan et al. 1992). According to AMAP (2000), 43 000 and 9500 t of Sovol were produced at Orgsteklo (1939–1990) and Orgsintez (1972–1993), respectively.

Sovtol (Soviet oil) has been characterised as a mixture of Sovol and trichlorobenzene (Ivanov and Sandell, 1992; AMAP, 2000). In particular, Sovtol-10 is a mixture of 90% Sovol and 10% trichlorobenzene (AMAP, 2000). Although other Sovtol-mixtures are known (Ivanov and Sandell, 1992), we assumed that all Sovtol contained 90% Sovol. According to AMAP (2000), a total of 32 000 t of Sovtol were produced at Orgsteklo (1939–1987) and another 25 000 t at Orgsintez (1972–1990). This results in an estimated total production of Sovol-based PCBs of 103 800 t. This figure corresponds well with the 100 000 t previously estimated by Ivanov and Sandell (1992).

Ivanov and Sandell (1992) also refer to another formulation produced in the former USSR, named 'Trichlorodiphenyl' (TCDP), stating that this was only a product name and that its chemical composition was fairly close to that of Aroclor 1242. We assume this to be the same formulation as TCB; Trichlorobiphenyl in AMAP (2000). This assumption seems reasonable in light of the fact that both sources list the same production period and use as a dielectric fluid. According to AMAP (2000), 70 000 t of TCB were produced at Orgsteklo during 1968–1990. This value is also used here, although it is considerably higher than the previous uncertain estimate of 25 000 t given by Ivanov and Sandell (1992).

The homologue composition of Sovol and TCB was taken from Ivanov and Sandell (1992) and is given in Table 2. For the congener production, we

assumed Sovol to be equal to Aroclor 1254 and TCB to be equal to Aroclor 1242.

3.1.3.7. China. PCBs were neither produced nor used in large amounts in China (Xu et al., 2000). According to Jiang et al. (1997), only 8000 t of PCB were produced in China during the 1960s and 1970s. The two principal technical formulations are said to be similar to Aroclor 1242 and Aroclor 1254 (Jiang et al., 1997). In order to estimate the homologue and congener production, we assumed that these two technical mixtures were produced in equal amounts and had the chemical composition of the above mentioned Aroclors.

3.1.3.8. France, Spain and Italy. The data from France, Spain and Italy include total production figures and detailed annual records for the years 1973–1984. Otherwise, only the factories and trade names, as well as a few data on the homologue and congener content of some of the technical mixtures were found in the literature (de Voogt and Brinkman, 1989; Kannan et al. 1992). As no data on annually produced amounts of various mixtures or constituents were available to us, the estimated production of individual homologues and congeners had to rely on the default homologue and congener compositions.

3.1.4. Uncertainties in the global production pattern

The methods applied to estimate production rates of individual homologues and congeners rest on a number of critical assumptions. The use of the default homologue and congener composition (e.g. for France, Spain and Italy) introduces uncertainties that are difficult to quantify. Obviously, the use of these default values implies that there were similarities in the production process between various producers, and implicitly, that there is a different propensity among the congeners to be formed during the production process. For some countries (e.g. China, USSR, Czechoslovakia), similarities have been noted in the chemical composition of technical mixtures from different producers. It thus seems reasonable to assume that there have indeed been similarities in the production process between different producers, resulting in similarities in the composition of the mixtures, at least at the homologue level (see, e.g. Takasuga et al. 1996). We are aware that there are substantial variations in the propensity of congeners to be formed during the manufacturing process (Frame, 1997). As the selected approach based on default compositions was designed to capture this variation, we only estimated production-weighted compositions based on technical mixtures that had been completely characterised (Schulz et al. 1989; Frame, 1997).

In any case, the available data make it difficult, if not impossible to quantify *all* of the uncertainties in a quantitative and meaningful way. For example, it is likely that the composition of the same technical mixtures varied, at least to some extent, from batch to batch (de Voogt and Brinkman, 1989; WHO, 1993). Secondly, the relative production rates of various mixtures were in many cases extrapolated based on reported data available for a few years only (e.g. Bayer AG).

However, the impact of the use of homologue and congener compositions can be addressed in a simplified manner. The same applies for the variability between Aroclor compositions determined with either ECD or MS systems (see Frame, 1997). Therefore, we tried to quantify and depict these two sources of uncertainty. Table 3 summarises how uncertainty was addressed for the production by various producers. Details are explained in the following.

At the *homologue level* (Table 3), the analytical variability (i.e. the difference between the homologue content of the Aroclors determined by ECD and MS) is estimated for the Aroclor production at Monsanto (USA). For most producers, the maximum and minimum homologue compositions are used to estimate some of the anticipated variations at the homologue level. Kanegafuchi (Japan) was treated as a special case, accepting the estimates for tetra- and penta-CBs as reported, and applying max/min compositions as determined specifically for this particular producer. The uncertainties at the homologue level for Bayer AG, Chemko, Orgsteklo, Orgsintez and Xi'an were not addressed because we anticipate that the assumptions made are of less quantitative importance than the variability in these compositions.

Table 3
Overview of the selected approach to estimate the uncertainties associated with the use of homologue and congener compositions as well as the variation in the characterised mixtures of Aroclors

	Analytical variability	Max/min compositions	Not addressed
Homologue level	Monsanto (USA)	Geneva Industries Kanegafuchi Mitsubishi Prodelec S.A. Cros Caffaro Monsanto (UK)	Bayer AG Chemko Orgsteklo Orgsintez Xi'an
Congener level	Monsanto (USA) Orgsteklo Orgsintez Xi'an	Geneva Industries Kanegafuchi Mitsubishi Prodelec S.A. Cros Caffaro Monsanto (UK) Chemko	Bayer AG

At the *congener level*, we used the analytical variability for Monsanto (USA) as well as for other producers for which the congeneric composition of the production was deduced from the Aroclors (Orgsteklo, Orgsintez and Xi'an). For most other companies, we used the maximum and minimum congener compositions multiplied with the corresponding max/min homologue compositions. The uncertainties at the congener level somehow represents a 'worst case' (max/max and min/min compositions). For Bayer AG, the uncertainty has not been addressed, due to lack of useful quantitative information for this purpose.

Fig. 2 presents results for the estimated total global production of individual homologues (A) and 22 selected congeners (B). The results indicate that of the 1324-kt total PCBs accounted for 566 kt (42.7%) can be attributed to the 22 selected congeners. Fig. 2 also indicates that tri-CBs have been the most important PCB homologue produced historically. Whereas there are theoretically 24 trichlorinated congeners, ranging from PCB-16 to PCB-39, a closer inspection reveals that approximately 50% of this homologue group can be attributed to only three congeners: PCB-18, PCB-28 and PCB-31. Similarly, the relative contribution of the other selected congeners to their respective homologue groups are 49% (Di-CBs), 24% (Tetra-CBs), 50% (Penta-CBs), 59% (Hexa-CBs), 22% (Hepta-CBs) and 43% (Octa-CBs). Table 4 presents the estimated global production rates of individual homologues for various time-periods (in percent). As can be seen from Table 4, the global homologue production pattern has changed over time. Notably, there was a reduction in the relative contribution of PCBs with seven or more chlorines over the last few decades that PCBs were produced. This is easily explained as the production of heavier technical mixtures decreased in the USA and West Germany in the last years of production (USA from 1970, West Germany from 1974) as a result of an increased environmental awareness towards the heavier (more persistent) homologues and congeners. For example, the estimated relative importance of Hexa-CB produced by Monsanto (USA) decreased by approximately 50% from the 1960s to the 1970s. Furthermore, the production of Hepta-CBs and Octa-CBs by Monsanto (USA) essentially ceased after 1973. This has profound influence on the applied homologue and congener compositions and their time dependence. Thus, Table 4 reflects the assumption that a similar decrease in the production of the more chlorinated

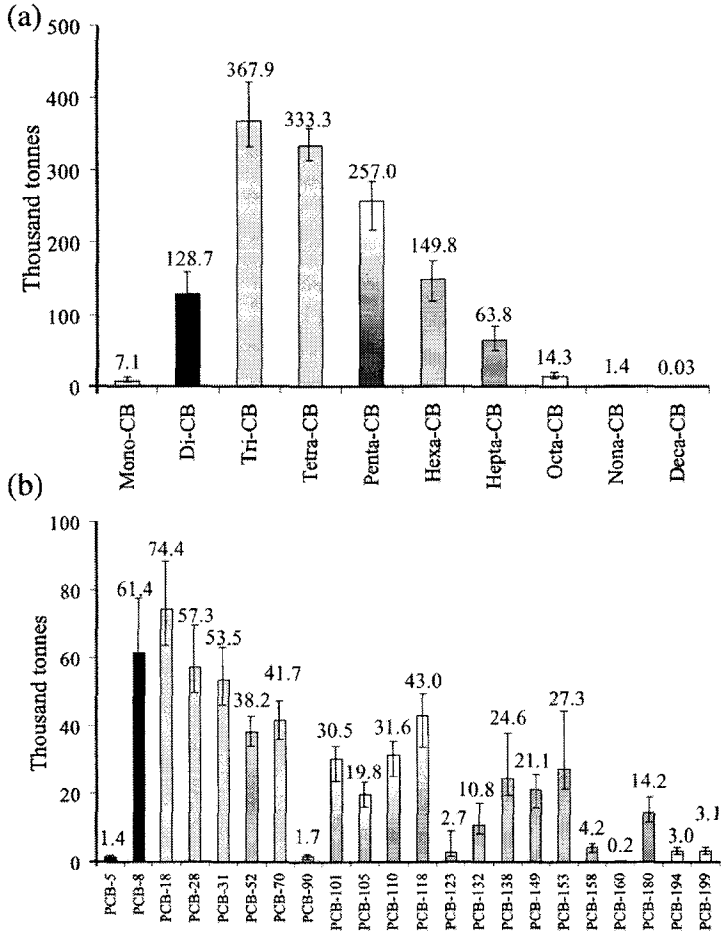

Fig. 2. Estimated global production of individual PCB homologues (A) and congeners (B) in thousands of tons.

congeners occurred worldwide around the same time. Unfortunately, this cannot be verified with the available information.

Fig. 3 presents results for the estimated temporal pattern in the global production of total PCBs (A), PCB-28 (B), PCB-52 (C), PCB-101 (D), PCB-118 (E), PCB-138 (F), PCB-153 (G), PCB-180 (H) from 1930 until 1993. The compiled data suggest a peak annual production of 75.5 kt total PCBs for the year 1970. Presumably, worldwide production of PCBs ended in 1993 when the production of Sovol ceased in Russia (AMAP, 2000). The results indicate further that the time trend of the production of individual congeners resembles that of the total PCBs. There are, however, notable exceptions when the uncertainties are taken into consideration. For the heavier congeners (Fig. 3F–H), the uncertainty is relatively high for the years after 1970. Again, this is a reflection of the use of the (*default*) homologue and congener compositions and their time dependence. As a result, the uncertainty for the later years of production increases. The negligible uncertainty depicted for individual congeners prior to 1955 are *not* due to the availability of more reliable data for this period. Rather, these historical data reflect that there were only a few producers worldwide and the production was dominated by Monsanto (USA) and Bayer (West Germany). The possibility to present any meaningful quantitative estimate of the uncertainties during that time period is considered limited.

Table 4
Estimated global production rates of individual homologues (in percent) and sum of all homologues (in kt) for various time-periods

Period	Mono-	Di-	Tri-	Tetra-	Penta-	Hexa-
1930–1934	0.3	8.6	25.0	24.5	16.0	14.3
1935–1939	0.3	8.2	24.1	24.4	17.4	14.6
1940–1944	0.3	7.2	21.1	24.2	22.0	15.5
1945–1949	0.3	7.2	21.1	24.2	22.0	15.5
1950–1954	0.3	7.2	21.1	24.2	22.1	15.5
1955–1959	0.5	7.5	21.5	23.5	21.4	15.3
1960–1964	0.7	8.3	23.4	24.2	20.5	13.8
1965–1969	0.8	10.3	28.7	26.9	18.2	9.4
1970–1974	0.8	11.1	32.0	24.9	17.5	9.4
1975–1979	0.2	10.3	29.8	24.9	21.3	10.7
1980–1984	<0.1	13.7	37.2	26.0	16.0	6.2
1985–1989	0	7.3	26.0	27.7	27.5	11.1
1990–1993	0	6.3	22.5	27.0	31.0	12.6

Period	Hepta-	Octa-	Nona-	Deca-	Sum homologues (kt)
1930–1934	9.1	2.1	0.2	<0.1	36.4
1935–1939	8.8	2.0	0.2	<0.1	37.8
1940–1944	7.8	1.7	0.2	<0.1	43.5
1945–1949	7.8	1.7	0.2	<0.1	43.5
1950–1954	7.7	1.7	0.2	<0.1	43.8
1955–1959	8.2	1.9	0.2	<0.1	97.7
1960–1964	7.3	1.7	0.2	<0.1	168.4
1965–1969	4.4	1.0	0.1	<0.1	285.6
1970–1974	3.4	0.8	<0.1	<0.1	274.7
1975–1979	2.3	0.4	<0.1	<0.1	169.5
1980–1984	0.7	0.1	<0.1	<0.1	86.6
1985–1989	0.5	0	0	0	29.2
1990–1993	0.6	0	0	0	6.8

3.2. Global consumption

The global consumption of PCBs has been estimated based on information about import, export and national consumption, as well as restrictions on imports of PCBs in various countries and regions.

In the absence of detailed information, which would facilitate an analysis for individual technical mixtures, we generally assumed that the import and consumption of individual congeners are determined by the production within representative countries or regions. This seems to be the only approach feasible with respect to the available data. Perhaps the most serious limitation in this methodology is that PCBs were exported as technical mixtures, rather than as individual congeners, as it is well-known that PCBs were sold according to the physical properties of the technical mixtures (WHO, 1993). Hence, some countries most likely had a relatively high import of one type of technical mixture (or product containing one type of a mixture). However, by treating groups of countries as closed markets, potential important regional variations in the spatial and temporal patterns of homologue and congener consumption can be addressed.

An overview of the method to address the global consumption pattern is given in Fig. 4, while details of the approach are discussed below.

3.2.1. OECD countries — consumption and export

Detailed data on imports and exports from OECD-countries, along with data on exports to OECD and non-OECD countries, are available for the period 1973–1980 from de Voogt and Brinkman (1989). Tatsukawa (1976) reported annual data on import and export for the entire period of

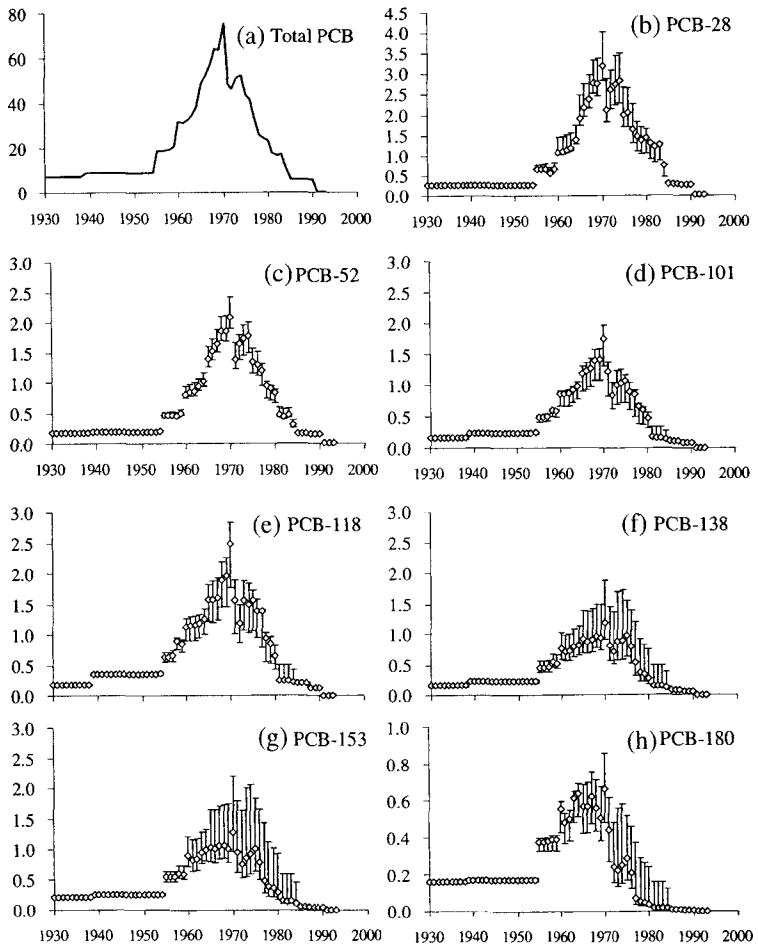

Fig. 3. Estimated temporal trend in the global production of total PCBs (A); PCB-28 (B); PCB-52 (C); PCB-101 (D); PCB-118 (E); PCB-138 (F); PCB-153 (G); and PCB-180 (H) in thousands of tons.

the PCB production in Japan. A key assumption for the national consumption within the OECD countries was that the OECD was a closed market. Whereas export from OECD was allowed to occur, import of PCBs into the OECD from non-OECD-countries was considered negligible. This should be a valid assumption in terms of quantitative importance, even though exceptions are likely. Furthermore, we assumed that prior to 1946, no export from producing countries occurred. For example, PCBs have been imported and used in Norway since approximately 1950, according to the national environmental protection authorities (SFT, 1996).

The annual national consumption of PCB-congeners in OECD-countries in the 1970s was estimated in the following way. de Voogt and Brinkman (1989) reported the annual exports from producing countries within the OECD to both OECD and non-OECD countries (see Fig. 4). The total annual export amount was thus divided into two sums, one for distribution to OECD countries and one for non-members. Next, we utilised information on annual imports of PCBs for specific countries within OECD for the same time-period (de Voogt and Brinkman, 1989). For all producing OECD-countries, there were data on imported amounts for most reference years of the given

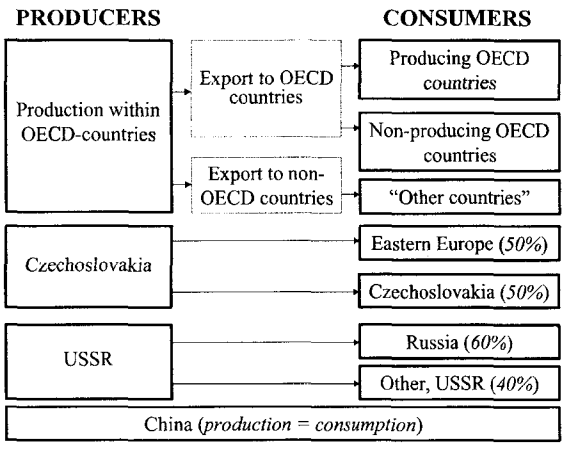

Fig. 4. Overview of the spatial distribution of global PCB consumption. Details of the approach are discussed in the text.

time-period, except for a few years for which we estimated the import with linear interpolation. Imports to all non-producing OECD-countries were based on annual reported imports whenever possible. The 'excess export amount' to OECD countries that could not be accounted for, was distributed to the remaining OECD countries based on gross domestic products (reference year 1983) (United Nations, 1994). An easy approach to evaluate this assumption would be to compare our estimates with independently derived estimates on national PCB consumption from e.g. national environmental agencies (see discussion later). Finally, for producing OECD-countries, we utilised either national consumption data *or* added the sum of import plus the amounts produced but not attributed to export, to estimate the national annual consumption.

For the years from 1946, up to the time when unifying statistical data became available, we had to make some simple assumptions, except for Japan where detailed data are available (Tatsukawa, 1976). We assumed that producing OECD-countries were exporting the same relative amounts of the total production as reported for the early 1970s. We used the gross domestic products of the OECD countries to distribute the estimated total export to OECD-countries. Canada was treated differently as its total cumulative import had been previously estimated to be approximately 40 kt (de Voogt and Brinkman, 1989). In our calculation we assumed a steady linear increase in the import from 1946 until 1972, withdrawing the amount imported to Canada in the 1970s.

Concerning the export to non-OECD countries (see Fig. 4), we assumed a linear increase in the fraction of export from 1946, until export-figures from OECD-producing countries to non-OECD countries became available for the early1970s.

This approach leads to uncertainties in the estimated national consumption data, and it is difficult to validate the results. However, the estimated data are comparable to some other independently derived national estimates. A national survey estimated the cumulative use in Austria to be between 2300 and 2800 t (Maderner and Hobiger, 1996), while our approach-which relies on GDP as a surrogate parameter-suggest a cumulative consumption of 3075 t. In a similar survey for Norway, the cumulative consumption has been estimated to be approximately 1230 t (SFT, 1996), while our data suggest 944 t.

3.2.2. Export from OECD to other countries

The amount of PCBs reported or estimated as being exported to non-OECD countries equals 148.3 kt or 11.2% of the total global production. We generally assumed that no PCBs were exported from OECD to China, the USSR or Eastern Europe. For the sake of simplicity, we further assumed no export to countries with a Gross Domestic Product (GDP) of less than US$ 1000 per capita or total GDP of less than one billion US$. The export to non-OECD countries includes 69 different countries. While the criteria to exclude some countries are somewhat arbitrary, these countries (i.e. approx. 70) would have accounted for a potential consumption of only 6.3% of the total export to non-OECD countries (or 0.7% of the total global production) — if included. This suggests that this simplification introduces minor uncertainties into the overall inventory at the scales of interest.

3.2.3. Eastern Europe

According to Šabata et al. (1993), approximately half of the amounts of PCBs produced in

Czechoslovakia was exported to other Eastern European countries. Hence, 50% of the annual Czechoslovakian production of PCBs was distributed among the countries of Eastern Europe, according to the Gross Domestic Product of the countries within the region. For comparison, the total export from the Czechoslovakia adds up to 21.5 kt or 1.6% of total global production.

3.2.4. Countries within the former Soviet Union

According to AMAP (2000), 60% of the Sovtol produced was used in Russia, and the rest in the former republics of the USSR. Similarly, 60% of the industrial capacitors containing PCBs were used in Russia. As a general assumption, we thus assumed that the former Soviet Union was a closed market where 60% of the total production was used in Russia. The remaining 40% were distributed among the other states of the former USSR according to Gross Domestic Product. Altogether, it is estimated that 173.8 kt or 13.1% of the global production have been used in the former Soviet Union.

3.2.5. China

Only a minor quantity of PCBs is reported to have been produced in China (8000 t or 0.6% of the total global production). We assume that all of the PCBs produced has been used within China.

Fig. 5 shows the estimated cumulative global consumption pattern for total PCBs, and includes estimates for 114 individual countries. It is estimated that USA has been responsible for as much as approximately 46% of the total historical global PCB consumption. Other major consuming countries include Russia (7.9%), Germany (7.1%), Japan (4.1%), France (4.1%), Canada (3.0%), Ukraine (2.4%), Spain (2.4%), Italy (2.1%) and UK (2.0%). The data at the national level should be interpreted with great caution. Particularly for the countries relying entirely on the assumptions related to GDP. It is, however, imperative to keep in mind that the overall objective of this study is to try to capture the overall spatial pattern of PCB consumption at a global scale. If the emphasis was at the national level, a different approach would be required.

In Fig. 5, the spatial distribution of the total historical national consumption is based on population densities within each country by use of the GEIA grid system of 1° by 1° (Dr Yi-Fan Li, Environment Canada). Population density is considered a suitable surrogate parameter, as the consumption of PCBs is generally linked with the use of electrical equipment. Overall, the results suggest that almost 97% of the intentionally produced PCBs have been used in the Northern Hemisphere. Furthermore, approximately 18% of the total have been used between 40 and 42° Northern latitude.

The results also show that there are temporal changes in the latitudinal distribution of PCB consumption. Fig. 6 shows how the latitudinal consumption patterns changed in time for total PCBs. As can be seen from this figure, the highest peak occurred in the 1960s (Fig. 6D). In the last period considered, the figure reflects the continuing consumption of PCBs in countries of the former Soviet Union (Fig. 6F).

The resulting dataset reveals that there are temporal and spatial variations in the homologue and congeneric consumption pattern. Fig. 7 exemplifies the regional differences in the historical consumption pattern for selected PCB congeners. The regions were selected in a way that is reflecting the method used to estimate the global consumption (see Fig. 4). We observe from Fig. 7 that PCB-101 and PCB-118 appear to be of limited significance in the consumption estimates for Eastern Europe as compared to the other regions. Furthermore, PCB-180 has been of relatively minor importance among the selected congeners in the former Soviet Union and China. However, these differences should be interpreted with care. For example, according to Takasuga et al. (1996), the technical mixture produced in Poland resembles Aroclor 1260. Hence, the estimates presented here for consumption in Poland are likely biased towards the lighter congeners as the production rates are lacking and hence not included in this inventory. The information may, however, be more reliable in other countries. For example, the available information would suggest that homologues with more than eight chlorines have not been used in the former Soviet Union (see also Table 2).

4. Final remarks

The global spatial and temporal consumption pattern of individual PCBs is considered essential

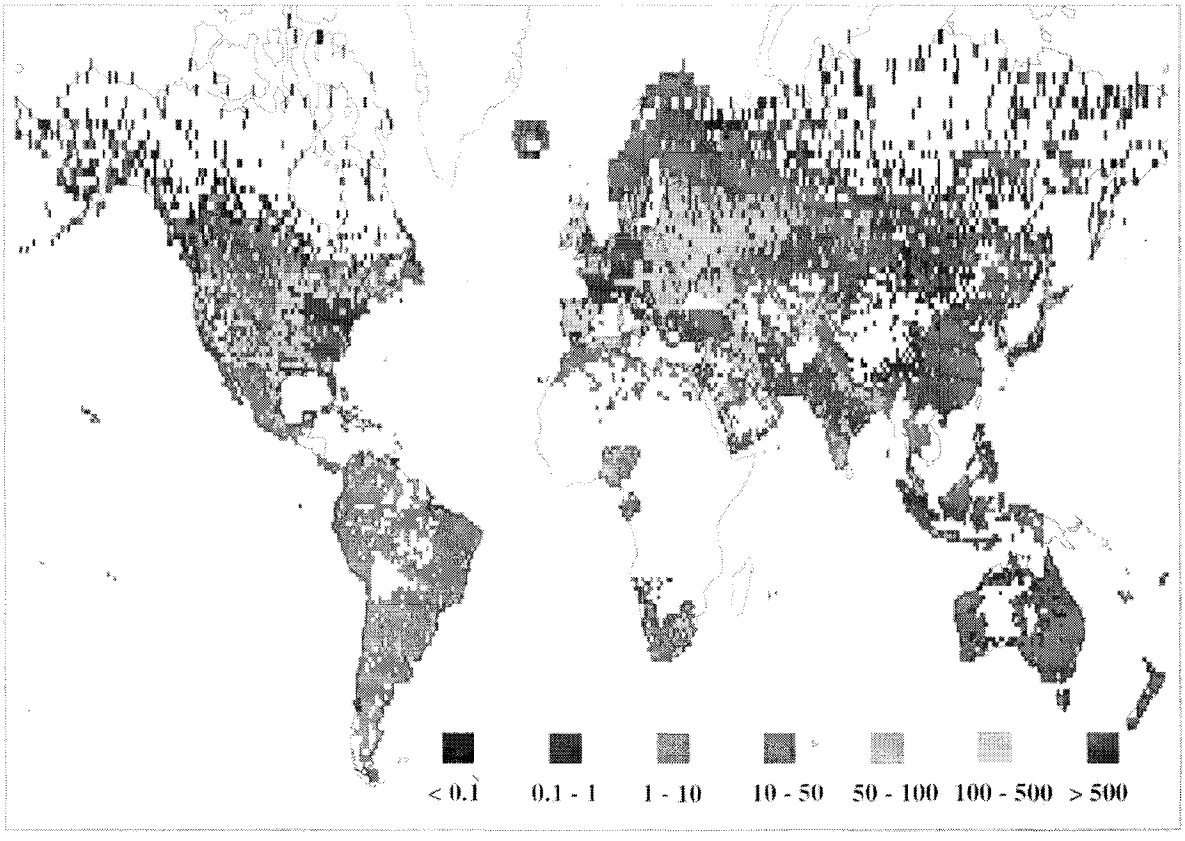

Fig. 5. Estimated cumulative global usage of PCBs (legends in t) with 1°×1° longitude and latitude resolution.

information for the interpretation of global PCB contamination patterns. Considering the large temporal and spatial scales of this approach, it is difficult to ensure that all relevant information has been considered. Indeed, we expect that for certain countries and years, more reliable data are available than those included in this study. Furthermore, only selected aspects of the involved uncertainties could be addressed here in a meaningful and quantitative way. We can presently only recognise other sources of uncertainty in a qualitative manner. For example, we are certain that there has been some production of PCBs in other countries, and that the compiled production data may be underestimated. Another source of uncertainty is that the estimated production of individual homologues and congeners for the first decades essentially remains unknown, but these data are obviously of less relative importance for current environmental levels. In spite of these uncertainties, we are confident that important aspects of the temporal and spatial pattern of global consumption are reflected in this inventory, although the uncertainties may be significant at a more detailed level, e.g. for single consuming countries, year and for some individual congeners. The availability of information for major producing companies and consuming countries around the time of their peak production indicates that the recent data are more accurate than the data from the past.

5. Additional information

Selected data from this study are available through internet as Microsoft Excel spreadsheets at www.nilu.no/projects/globalpcb/.

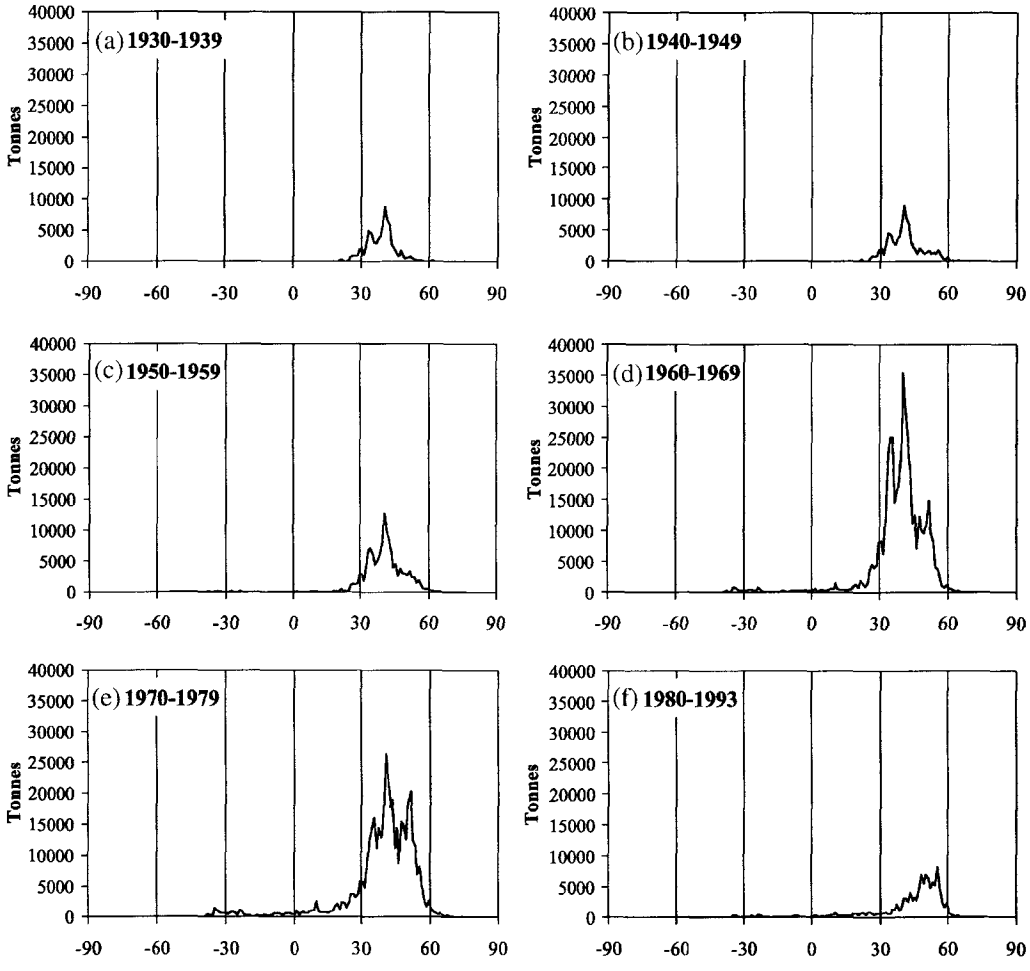

Fig. 6. Global consumption of total PCBs for six different time-periods (by latitude).

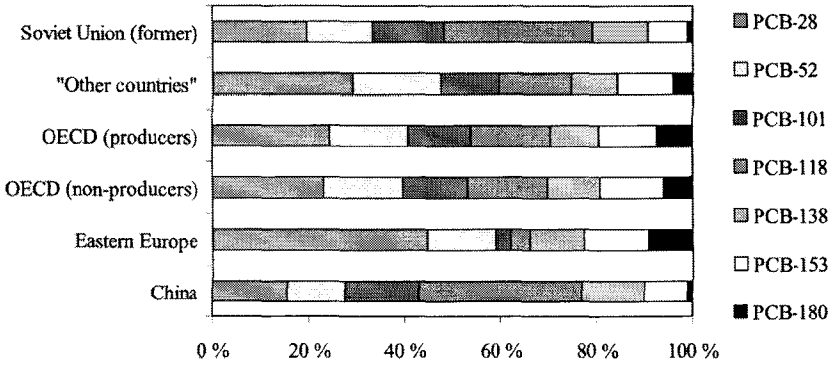

Fig. 7. Regional differences in the cumulative consumption of selected PCB congeners (in percent).

Acknowledgments

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