Annex Section (a) – Sources of PeCB

This annex contains data and references to support the information provided in Section (a) of the WCC Submission for Pentachlorobenzene (PeCB)

Extracted from Bailey, B., 2007. Pentachlorobenzene – Sources, environmental fate and risk characterisation, Euro Chlor Science Dossier (in press).

1   PeCB Sources in the environment

No global inventory of possible emissions of PeCB to the environment was found. However, in Canada total releases of PeCB have been estimated to be 41.8 kg/y. PeCB from backyard trash burning is estimated to be 21.93 kg/y, pentachlorophenol treated wood, 2.34 kg/y, pesticide use 6.2 kg/y, dielectric fluid 5.6 kg/y, municipal solid waste incineration 2.36 kg/y, hazardous waste incineration 1.84 kg/y, magnesium production 1.53 kg/y and solvent use 0.04 kg/y (Environment Canada 2005). No estimates of PeCB emissions in the remainder of the world were found. Thus, the following factors for estimating global PeCB emissions were developed for this report

1.1   Industrial uses of PeCB

PeCB is not known to have any commercial uses at present (Beck 1986; Environment Canada 1993). However, in the past, PeCB was one component of a chlorobenzenes mixture used to reduce the viscosity of PCB products employed for heat transfer (Environment Canada 1993; King et al. 2003). PeCB has also been used in a chlorobenzenes mixture with PCBs in electrical equipment (Environment Canada 2005). PCBs are still in use in some old electrical equipment in North America and Europe so that there is a small potential for release of PeCB from this source (AMAP 2000; Environment Canada 2003). It can be presumed that some PCBs are also still in use elsewhere in the world and some fraction of them contain PeCB.

PCBs are being taken out of service in many countries of the world so that any related PeCB emissions are expected to be decreasing with time. Global releases of PCBs have been estimated to be approximately 40 metric tons in 2000 (Breivik et al. 2002). Breivik noted a high level of uncertainty in the emissions estimate, plus and minus a factor of 10. Breivik's estimate of Canadian PCB emissions was 0.6 metric tons per year. Assuming the same ratio of PeCB emission to PCB emission, the global emission of PeCB from use with PCBs can be calculated:

$$((5.6 \text{ kg/y PeCB})/(0.6 \text{ t/y PCB}))*(40 \text{ t/y PCB}) = 373 \text{ kg/y}$$

PeCB was used in the past as an intermediate in manufacture of pentachloronitrobenzene (quintozene) (van de Plassche et al. 2001). However, quintozene is now made by chlorination of nitrobenzene (Feiler 2001). PeCB may also have been used in the past as a fungicide and flame retardant (van de Plassche et al. 2001).

1.2   Byproduct formation of PeCB

The U. S. Toxics Release Inventory (TRI) includes PeCB. A total of 20 industrial facilities are listed as emitting or transferring 2533 pounds (1151 kg) of PeCB in 2004 (USEPA 2006). The industries represented are chemical, waste treatment, and coal burning electric power. Total reported air emissions were 220 pounds (100 kg) and water emissions of 17 pounds (7.8 kg) (USEPA 2006). In industrial chlorination reactions it is possible to produce PeCB as a byproduct and it probably accounts for some of the emissions reported in the TRI. There are other processes which produce a variety of chlorinated aromatics that may contribute PeCB even if PeCB has not been explicitly detected and reported yet. TRI includes only industrial facilities handling relatively large amounts of chemicals so that additional emissions are expected.

One way to estimate global emissions from the global chemical and related industry sectors covered by the TRI program in the USA is to simply multiply by the ratio of world gross domestic product (GDP) to the US GDP. Since the USA accounts for about 25% of world GDP, multiplying the TRI reported emissions to air and water by 4 leads to estimated global

emissions of 431 kg/y. This value is uncertain because of both the uncertainty in the US emissions and waste handling practices in developing industrial countries. North American emissions of PeCB were greater in the past due to less stringent waste handling regulations as shown by monitoring studies described in Section 4.

Global emissions from chlorinated solvent use and pesticide use have been estimated by scaling from the Canadian emissions reported by Environment Canada (Environment Canada 2005). In round numbers, the Canadian GDP is about 2.5% of the global GDP. Therefore, multiplying the PeCB emissions from the use of chlorinated solvents, 0.04 kg/y, by 40 gives an estimate of 1.6 kg/y for global emissions from chlorinated solvents. Emissions from pesticide and pentachlorophenol use are calculated similarly:

$$(6.2 + 2.34 \text{ kg/y})*(40) = 341.6 \text{ kg/y}$$

The emission of PeCB has been reported from the use of hexachloroethane (HCE) to remove dissolved hydrogen from molten aluminum in foundries (Westberg et al. 1997). They reported an emission rate of 310 μg PeCB/g HCE used at a typical HCE application rate. While the use of HCE for degassing aluminum in foundries is not widespread in the USA, several small foundries were reported to use HCE (Streeter 1998). Because HCE is easier to use than gaseous chlorine for degassing aluminum, it is probably used in some of the less developed countries as well as in smaller facilities in developed countries. In 1998 it was reported that about 3,500,000 kg/y HCE was used in this application (Scottish Chemical 1998). Using the Westberg et al. factor, this could result in the emission of:

$$(310 \text{ } \mu\text{g/g})*(3,500,000 \text{ kg/y})*(10^{-9} \text{ kg/}\mu\text{g})*(1000\text{g/kg}) = 1100 \text{ kg/y PeCB}.$$

Vogelgesang also reported the release of PeCB and other compounds from degassing of aluminum (Vogelgesang 1986). PeCB emissions from an aluminum smelter using a sodium/potassium chloride flux were reduced from 18.05 μg/m^3 to 10.34 μg/m^3 by installation of a baghouse filter (Aittola et al. 1996). The other chlorobenzenes and chlorophenols were reduced by a similar fraction. In contrast, the filter reduced the polychlorinated dioxins and furans by 98-99%.

There are several processes for production of metals involving the treatment of ores with carbon and chlorine to yield a soluble or volatile compound which can be purified. Carbochlorination of magnesium oxide, heating MgO with coke to 700-800 °C in a chlorine atmosphere, yields $MgCl_2$ and CO along with traces of PeCB, HCB and dioxins/furans and other chlorinated compounds (Knutzen and Oehme 1989). Knutzen and Oehme estimated that about 50 kg/year PeCB along with up to 300 kg HCB may have been emitted to the Frierfjord, Norway during the 1980s (Knutzen and Oehme 1989). Norwegian regulations called for dramatic reduction in dioxin/furan emissions in the 1990s so that it may be assumed that PeCB emissions have decreased also (although not necessarily in direct proportion). Recovery of copper from a slag by roasting it with coal and sodium chloride produced a variety of highly chlorinated organics (Doering et al. 1992). The residue from this process was an attractive red gravel which was used in landscaping. Concentrations of PeCB in this red slag ranged from 0.6 to 1200 μg/kg and HCB ranged from 5.3 to 11000 μg/kg in samples. Vogelgesang reported release of PeCB and other chlorinated compounds from the production of niobium and tantalum (Vogelgesang 1986). The author of this report, Bailey, speculates that other processes which use chlorine and carbon at high temperatures may produce PeCB, such as the chloride process to produce TiO_2 pigment. No quantitative estimates are provided because there is no quantitative information on which to base them.

1.3 Combustion formation of PeCB

The observation of PeCB as a trace product of incomplete combustion has been widely reported. Nearly all fuels contain some chlorine, especially biomass and waste. The yield of PeCB from combustion of different fuels under different conditions has been reported to vary widely. The goal in this section is to extract PeCB yield factors from the various research studies and apply these factors to combustion processes for estimation of global PeCB emissions from combustion.

1.3.1 Waste combustion

Open burning of household waste and the resulting emissions of PeCB averaged 0.076 (range 0.033-0.162) mg/kg mass actually burned or a factor of 7.6×10^{-8} kg/kg (Lemieux 1997). The variation between duplicate burns in this small project prevent any conclusions from the two different waste mixtures used and illustrate the uncertainty in the emission factor. This study was designed to represent uncontrolled combustion of solid waste in a perforated barrel. It probably applies to much of the uncontrolled burning of trash which takes place in piles all over the world as well as landfill fires.

Emissions of PeCB and other chlorobenzenes during incineration have been studied as a potential way to estimate emissions of chlorodioxins and furans (PCDD/F) with simpler analytical technology. The use of PeCB for monitoring has been suggested by Kato and Urano who found a correlation between PeCB concentrations and the international toxic equivalent factor (I-TEQ) in 5 different operating waste disposal plants in Japan (Kato and Urano 2001). They found that under normal operating conditions PeCB correlated with I-TEQ within a factor of 3.

Other workers have also reported correlations between the emission of PeCB and polychlorodibenzodioxins and furans (PCDD/F) (Oberg and Bergstrom 1987; Kaune et al. 1994). Aittola et al. pointed out that a filter that removed over 90% of the PCDD/F, removed only about 50% of PeCB and HCB from an aluminum smelter (Aittola et al. 1996). Thus reductions in PCDD/F emissions may not lead to equal reductions in PeCB emissions.

The reported yields of PeCB to HCB formed in different combustion processes vary widely, depending on combustion conditions and the presence (or not) of catalytic materials. For example, in a fluidized bed solid waste incinerator the yield of total chlorobenzenes and PCDDs increased 20 fold as metals accumulated in the sand, while the PeCB/HCB ratio was approximately the same (Akimoto et al. 1997). Because combustion conditions, for example, oxygen, time and temperature, seem to dominate in most situations, the influence of fuel composition, concentration of chlorine beyond a minimum concentration, on the PeCB emissions is only one of several factors.

Many of the studies of trace products of incomplete combustion report in terms of mass per volume of stack gas which is what the regulations require. To convert to a yield of PeCB in terms of mass of waste burned, a gas flow of 7,000 m^3/metric ton has been used (Carpenter et al. 1986). Combustion experiments are difficult to control and the PeCB from nominally duplicate runs varied by factors of 2 to 5 in those studies where replicates were reported. The variety of parameters studied also led to wide variation in the PeCB yields. Table 3.1 has only average PeCB yields and illustrates some of the variation reported. The yield of PeCB is certainly affected by poor combustion conditions. Another significant factor in the overall yield of chlorinated aromatics is the presence of solid surfaces, especially if metals are present. One conclusion from all this work is that PeCB is probably emitted from all combustion processes of fuels containing any chlorine as organochlorine or chloride.

Table 3.1. Yields of PeCB in various waste incineration experiments. The results of different experiments in each study have been averaged. PeCB concentrations have been converted to emission factors by multiplying by 7000 m^3 of stack gas per ton of fuel.

PeCB Emission Concentration	PeCB Emission Factor	Experimental	Study
0.91 μg/std m^3	6.4 mg/ton	Lab fluidized bed	(Wikstrom et al. 1999)
39 μg/std m^3	273 mg/ton	City incinerator	(Tiernan et al. 1983)
1 μg/std m^3	7 mg/ton	24 city incinerators	(Kato and Urano 2001)
23 μg/std m^3	161 mg/ton	Lab fluidized bed	(Fängmark et al. 1993)
12 μg/std m^3	84 mg/ton	Lab fluidized bed	(Fängmark et al. 1994)
0.42 μg/std m^3	2.9 mg/ton	City incinerator	(Jay and Stieglitz 1995)
25 μg/std m^3	175 mg/ton	City fluidized bed	(Akimoto et al. 1997)
2 μg/std m^3	14 mg/ton	Hazardous waste incinerator	(Oberg et al. 1985)
Special purpose experiments not used in waste combustion averaging.			
87 ng/std m^3	0.6 mg/ton	Wood burner	(Zimmerman et al. 2001)
	969 mg/ton	Pure PVC in pilot plant burner	(Ahling et al. 1978)
	300 mg/ton	Pure PVC in lab burner	(Kim et al. 2004)

The range of PeCB yields above extends over a factor of 100 from experiments designed to clarify the processes taking place during municipal incineration. The geometric mean of the emission rates from the waste combustion experiments above, except the Tiernan 1983 study, is about 25 mg/ton. For the purpose of an order of magnitude estimate of PeCB emissions, the emission factor for municipal and hazardous waste incineration will be 25 mg/ton of waste or 2.5×10^{-8} kg/kg. The total amount of waste incinerated with the low emission factor above is estimated to be about 200 million tons per year which could yield PeCB.

$(2.5 \times 10^{-8})*(200 \times 10^6 \text{ tons}) = 5 \text{ tons} = 5000 \text{ kg/y PeCB emitted.}$

In most of the developing world trash is burned casually to reduce its volume and attraction of vermin. The production of solid waste is estimated to be about 0.4 kg per capita-day (Zurbruegg 2003). For this report it is estimated that about half of the solid waste is burned. Thus a population of 5×10^9 people would generate 2×10^9 kg/day of total solid waste. Using the average PeCB emission factor from Lemieux, see above (Lemieux 1997), the yield of PeCB on an annual basis would be:

$(2 \times 10^9 \text{ kg/day})*(365 \text{ d/y})*(0.5 \text{ burned})*(7.6 \times 10^{-8}) = 27,740 \text{ kg/y}$

1.3.2   Coal combustion

Coal use in 2005 was estimated to be 4990 million metric tons (World Coal Institute 2006). Coal typically contains about 0.1% chlorine. No information on PeCB emissions from coal combustion was found. However 0.07 µg/std m^3 HCB has been reported from coal combustion in a lab experiment (Oberg and Bergstrom 1985). Oberg and Bergstrom did not report the conditions of combustion but simply reported "satisfactory and fully comparable". In contrast, HCB was not detected in a series of coal fired utility boilers tested in the USA (USEPA 1998). Because a sizable fraction of coal is burned in small domestic units, some PeCB emissions are expected. The references cited above generally reported several times more PeCB than HCB from combustion processes with low concentrations of chlorine. For the purposes of this report, half of the coal used is assumed to be burned domestically and the reported HCB emission concentration from Oberg and Bergstrom will be multiplied by 5. The PeCB emission factor is calculated:

$(0.07 \text{ µg/std m}^3)*(7000 \text{ m}^3/\text{ton})*(5) = 2450 \text{ µg/ton or } 2.45 \times 10^{-9} \text{ kg/kg PeCB per kg coal.}$
Multiplying this factor by coal burned in small scale combustion yields:

$(2.45 \times 10^{-9} \text{ kg/kg})*(0.5)*(4990 \times 10^9 \text{ kg}) = 6113 \text{ kg PeCB}$

1.3.3 Biomass combustion

Total biomass burning in the 1990s was estimated to be 3716 million metric tons of carbon with a chlorine content ranging from 2.4 mg/kg (tulip trees in Ohio, USA) to 9000 mg/kg (citrus leaves in Southern California) (Lobert et al. 1999). The only emission factor for PeCB from burning biomass found was that of Zimmerman where waste wood was burned in a high temperature incinerator (Zimmerman et al. 2001). No information on PeCB emissions from smoldering combustion or other combustion conditions was found. However, Gullet and coworkers have reported a series of studies on PCDD and PCDF emissions, expressed as international toxic equivalents (I-TEQ), from combustion of biomass under conditions designed to simulate agricultural burning and forest fires (Gullet and Touati 2003a; Gullet and Touati 2003b; Gullet et al. 2006). Table 3.2 summarizes the I-TEQ emissions from the different studies.

Table 3.2. Comparison of emission factors from biomass combustion studies.

Substance	I-TEQ ng/kg C emission factor	PeCB ng/kg C emission factor	Reference
Wood	0.8*	1200 ng/kg C	Zimmerman et al. 2001
Sugarcane	1.7-25**		Gullet et al. 2006
Forest fire	4-30		Gullet and Touati 2003a
Straw	~1		Gullet and Touati 2003b

* Emissions converted from concentration (ng/m^3) to emission factor (ng/kg C) by multiplying by 7000 m^3 flue gas per ton of wood burned, and assumes wood to be 50% carbon.
** The very high emission factor, 253 ng TEQ/kg C from Hawaiian sugarcane, was not used because it seems to be an outlier.

Several researchers have noted a correlation between the emission of PeCB and PCDD/Fs from waste combustion (Oberg and Bergstrom 1985; Kato and Urano 2001). Zimmerman et al. reported the average emission of PeCB was 1475 times the average I-TEQ emission in their wood burning experiment (87 ng/m^3/0.059 ng/m^3 = 1475). This ratio of PeCB to I-TEQ is within the range, 500 to 2500 ng PeCB per ng of TEQ, reported in the studies summarized in Table 3.1, above. Based on the I-TEQ emissions from uncontrolled biomass burning in Table 3.2, it is reasonable to estimate the PeCB emission factor for most biomass burning is somewhat greater than that reported by Zimmerman et al. For the purposes of this report, developing an approximate global emission inventory for PeCB, the average emission PCDD/Fs (I-TEQ) from combustion of biomass will be taken as 8 ng/kg C burned. Using the PeCB/I-TEQ ratio from Zimmerman the emission factor to be used for PeCB is 11,800 ng/kg C burned. Multiplying this factor by the amount of biomass burned yields an emissions estimate.

$$(11{,}800 \text{ ng/kg})*(3716 \times 10^6 \text{ ton})*(1000 \text{ kg/ton}) = 4.39 \times 10^{16} \text{ ng PeCB}$$
$$= 43{,}900 \text{ kg PeCB}$$

1.4 PeCB from degradation of other chemicals in the environment

Dechlorination processes for HCB have been studied in the laboratory. HCB was photodechlorinated to PeCB in surfactant micelles (Chu et al. 2002). The PeCB thus formed was subsequently dechlorinated at about the same rate as HCB. Therefore a negligible net production of PeCB is expected from this environmental process. Indirect photolysis of HCB in arctic lake water was reported to be facilitated by dissolved organic matter; however there was no mention of the products (Grannas et al. 2003).

Anaerobic biological dechlorination of HCB to PeCB followed by dechlorination of PeCB has been reported by many workers (Tiedje et al. 1987; Fathepure et al. 1988; Pardue et al. 1993; Ramanand et al. 1993; Beurskens et al. 1994; Pavlostathis and Prytula 2000). However, the dechlorination rate of PeCB is reported to be faster than that of HCB so that PeCB does not accumulate but is further dechlorinated to tetrachlorobenzenes and lower chlorobenzenes. Thus anaerobic biological dechlorination of HCB is not expected to lead to a net accumulation of PeCB in the environment.

Pentachloronitrobenzene (PCNB) has been reported to degrade in soil forming a small yield of PeCB along with much higher yields of other related compounds (Beck and Hansen 1974) . PCNB has also been reported to be photolyzed to PeCB along with other related compounds

(Crosby and Hamadmad 1971). Global agricultural use of PCNB in the 1990s was reported to be 880,590 kg (Landell Mills Market Research 1996). Thus there is the potential for the release of some PeCB. PeCB from this source was not included in the summary of PeCB sources, Table 3.3, because the available information did not allow calculation of the potential quantity.

1.5   PeCB global emissions inventory
Using the factors and logic described above, the estimated global emissions of PeCB around the year 2000 are summarized in Table 3.3, representing a total of about 85,000 kg/y. As described above there is considerable uncertainty about the size of each of these estimated PeCB emissions, potentially an order of magnitude. The global emissions are clearly dominated by combustion sources. Of all sources, combustion of biomass (43,900 kg/y), combustion of solid waste (32,740 kg/y) and combustion of coal (6113 kg/y) represent the three largest sources. Industrial sources are relatively minor and improvements in industrial practices have probably led to significant reductions in environmental concentrations of PeCB.

Table 3.2. Summary of estimated annual global emissions of PeCB.	
PCB use losses	373 kg
Chlorinated solvents	<2 kg
Pesticide use	342 kg
Chemical manufacturing and waste disposal	431 kg
Aluminum casting	1100 kg
Combustion of solid waste	32,740 kg
Combustion of coal	6113 kg
Combustion of biomass	43,900 kg
Total annual emission	85,001 kg

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