

Potential risk of remaining PCBs and management strategies in Europe*

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ABSTRACT

Polychlorinated biphenyls (PCBs) are persistent hazardous chemicals and are now out of production worldwide. Although over 20 years have passed since the Helsinki Convention promoted the elimination of PCB use, it is difficult to put an end to PCB emissions in a short time because PCBs have a long lifetime in the environment and a major portion of PCBs are emitted from electrical equipment manufactured before the

Helsinki Convention but still used today. PCBs have an impact on human sperm integrity in the European male population; furthermore, Arctic ecosystems such as those supporting polar bears and seals are exposed to risks due to the long-distance transport of PCBs. Most European countries have no proper facilities for PCB treatment. After analyzing extensive information about PCB treatments, it is concluded that the thermal process is technically sound and cost-effective. This study contributes to the promotion of treatments in most European countries storing PCBs with the aim of pursuing a sustainable future.

INTRODUCTION

Polychlorinated biphenyls (PCBs) consist of a mixture of homologous and/or isomeric chlorinated biphenyls. Theoretically, 209 compounds are possible, with various degrees of chlorination between 21% and 68%. PCBs with an average content of 42 to 54% chlorine are most common in terms of production and use (Munch and Axenfeld 1999). The toxicity of individual PCB congeners is largely governed by the number and position of the chlorines on the biphenyl nucleus: Figure 1 shows two examples to clarify the above description.

Chlorines in both *para* positions (4 and 4') and at least 2 chlorines in the *meta* positions (3, 5, 3', 5') are considered to be dioxin-like and are particularly toxic (see Figure 1B) (Ahlborg et al. 1994; US Environmental Protection Agency 1987).

Of special ecological interest are the congeners: 28, 52, 101, 118, 138, 153, 180, non-ortho 77, 126, 169 and mono-ortho 105, 118 (Munch and Axenfeld 1999). There are various sources of PCB emission (Full 2001): (i) evaporation of PCBs used in the past in open systems (products and buildings), (ii) evaporation of spilled or leaked PCBs from equipment containing PCBs still in use, (iii) evaporation from landfills or incinerators, (iv) evaporation of PCBs from improper disposal of PCB-containing products, (v) emissions of PCBs from engines and furnaces burning liquid or gaseous fuels containing PCBs or

contaminated with them, (vi) emissions of PCBs from open burning or incomplete incineration of waste, (vii) emissions of PCBs from small capacitors in household appliances, and (viii) evaporation of PCBs from waste oil recycling.

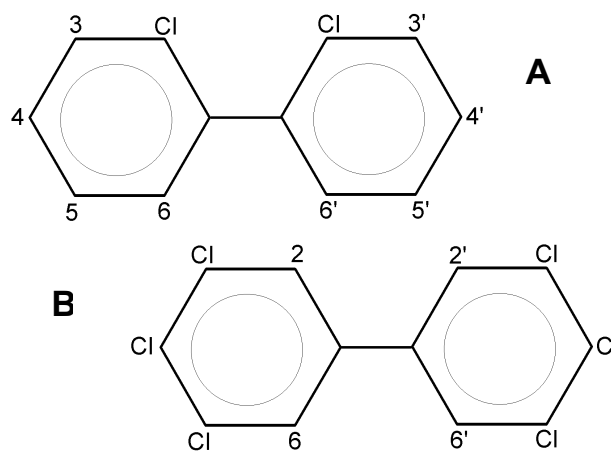


Figure 1. Basic PCB structure and toxicity. A – less toxic structure; B – more toxic structure (adapted from Ahlborg et al. 1994; US Environmental Protection Agency 1987).

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The Helsinki Convention promoting PCB elimination was adopted in 1985, so PCBs are basically out of production worldwide. However, world production (excluding the Soviet Union) totaled 1.5 million tons from 1929 (the beginning of their commercial use) to 1989 (UNEP 1999). Historical emissions of PCBs in Europe are shown in Figure 2.

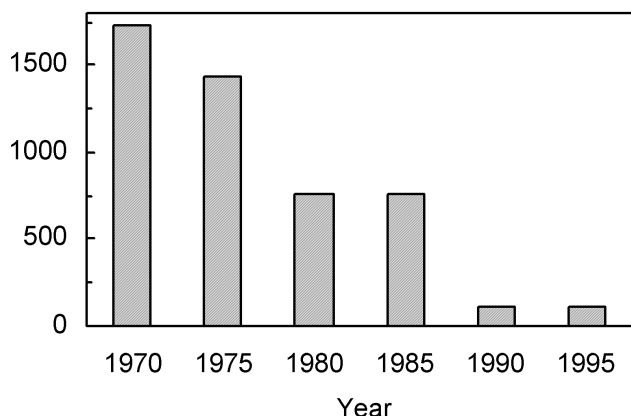
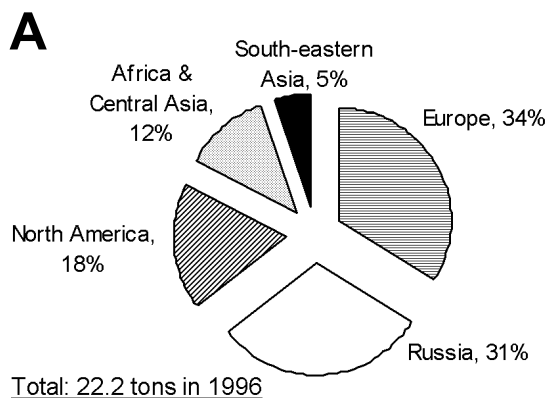


Figure 2. Historic emissions of PCBs in Europe (redrawn from Munch and Axenfeld 1999).

Figure 2 shows a dramatic decrease in PCB emissions, but European countries still have large quantities of PCBs, and the current quantities of liquid PCBs and solid PCBs are estimated at 200 thousand tons and 400 thousand tons, respectively (SBC 2003). It is very difficult to stop PCB emissions within a short timeframe by legal or technical measures because of the long lifetimes of the products containing PCBs (10-30 years) and their use in small capacitors which are usually disposed of with garbage (Munch and Axenfeld 1999).



MOTIVATION BEHIND THE RESEARCH

The European Commission decided to send reasoned opinions (second warning letters) to Spain, France, Greece, Ireland, Italy, Luxembourg, Portugal and the United Kingdom for their failure to submit information to the Commission as required under the Directive (No. 96/95/EU) on PCBs, because the eight Member States did not send inventories and plans on their handling and disposal of PCBs to the Commission despite an earlier warning (Wildlife News 2000). It may be difficult to collect current information because over 20 years have passed since the Helsinki Convention promoting the elimination of PCB use. It should be noted that Europe is the greatest emitter of PCBs in the northern hemisphere (Figure 3A) and the atmospheric concentration of PCBs is higher there than in other regions (Figure 3B).

According to a comment by the European Environment Commission (Wildlife News 2000), strict control of PCBs is essential. The timely implementation under the Directive is an integral part of the controls that have been put in place at the community level. The aim of the directive is to streamline the legislation of the member states on the decontamination (or disposal) of equipment containing PCBs and the disposal of used PCBs in order to eliminate them "completely". There is doubt as to whether the member states really strive to eliminate PCBs completely. This paper discusses (i) the present situation of PCBs, (ii) potential capacity of PCB elimination, and (iii) technologies available to European countries.

EFFECTS OF PCBs ON ECOSYSTEMS

It is reported that sea bass (*Dicentrarchus labrax*) are subject to the accumulative effect of PCBs in Ria de Aveiro, Portugal (Antunes et al. 2001): PCBs ranged between 407 ng · g⁻¹ lipid weight (lw) for octopus and 22,287 ng · g⁻¹ lw for bass in the

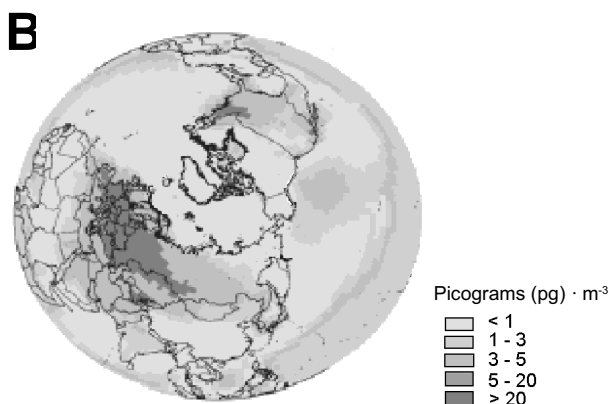


Figure 3. PCB-180 in the northern hemisphere in 1996 (adapted from Shatalov et al. 2003). A – contribution of PCB-118 to the northern-hemispheric emission by region; B – atmospheric concentration of PCB-118 (note: PCB-118 is an indicator PCB congener (2,2',3,4,4',5,5' heptachlorobiphenyl)).

Gulf of Naples, Italy (Domingo and Bocio 2007). In another measurement (the Huelva coast in Spain), the highest and lowest PCB mean values corresponded to sardine and prawn: $23,787 \text{ pg} \cdot \text{g}^{-1}$ wet weight (ww) and $861 \text{ pg} \cdot \text{g}^{-1}$ ww, respectively (Domingo and Bocio 2007). A mass balance of PCBs (e.g. UK) showed that soil retained larger amount of PCBs than other compartments did (air, freshwater, sediment, or biota) (Harrad et al. 1994). Soil is today the greatest source (90%) of PCBs to the atmosphere through re-circulation (Harrad et al. 1994), and the major portion of PCBs entering freshwater ecosystems come from the atmosphere (Full 2001).

Long-distance transport of PCBs

The more volatile PCBs are moving toward equilibrium with the organic matter burden of the soil compartment on a European regional scale, while the distribution of the stickier (heavier) homologues appears to still be primarily influenced by their preferential deposition closer to source areas. The relative concentration of the tri- and tetra-PCBs increases with latitude, while that of the hepta- and octa-PCBs decreases, consistently with the global fractionation theory (Meijer et al. 2002). Persistent chemicals in Arctic areas should be subject to more stringent factors than in temperate regions (Wania and Mackay 1999): this is principally because the climatic conditions tend to affect degradation rates and some persistent organic pollutants (e.g. PCBs) may thus experience an enhanced lifetime in cold climates (reviewed by Breivik et al. (2002). Based on monitoring data (AMAP 2003), the polar regions appear to be an environmental sink for PCBs. Figure 4 shows PCB levels in Arctic animals.

PCB levels measured in bear fat are approximately five times higher than in blood from the same animals (AMAP 2003). High levels of PCBs (and Dichlorodiphenyltrichloroethanes – DDTs) in polar bears are observed from the east coast of Greenland and around Svalbard. Recent studies have revealed even higher levels in polar bears from Franz Josef Land and the Kara Sea, with decreasing trends eastwards and westwards from this region (AMAP 2003).

There have been several suggestions to explain why some polar bear populations have much higher PCB levels than others. These include proximity to sources and contaminant transport by ice from source regions. New information about what polar bears eat might shed more light on the issue. Most polar bears feed on ringed seal, especially the blubber. However, east of Svalbard, an unusually high number of bears feed on harp seals. These seals migrate from the Russian White Sea and may serve as a biological pathway from Russia to Svalbard. Moreover, blubber from adult harp seals has higher concentrations of persistent organic pollutants (POPs) during the molting season in June, compared with ringed seals sampled at the same time. The data suggest that the season, availability, and biological condition of polar bear prey may play an important role in biomagnification of PCBs in the marine food web (Kikuchi et al. 2005). Data were collected from 81 polar bears on Svalbard (near the Russian economic zone) in order to study the relationships between PCBs and thyroid hormones (THs) (Braathen et al. 2004). Alterations in THs have behavioural and neurological consequences throughout the bears' life cycle (Sher et al. 1998), and TH regulation is important for maintaining normal body temperature (McNabb 1992).

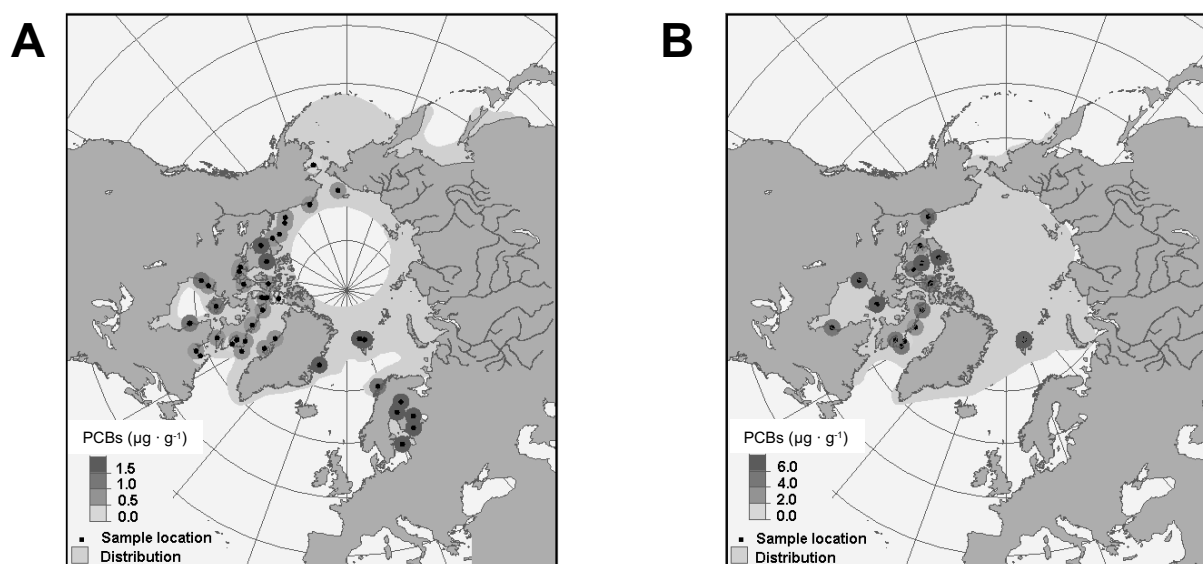


Figure 4. PCB levels in ringed seals (blubber) and polar bears (fat). A – PCBs in ringed seals; B – PCBs in polar bears (redrawn from AMAP 2003).

Effects of PCBs on European people

PCBs and dichlorodiphenyldichloroethylene (*p,p'*-DDE) biologically accumulate and are concentrated by up to a thousand times compared with background levels, particularly in high rank predators of the food chain, man included (Longnecker et al. 1997). Thus, these compounds are still detected in human blood, fat tissue and breast milk worldwide (Longnecker et al. 1997). Studies on wildlife and laboratory animals have shown that these compounds have adverse effects on the reproductive and endocrine functions (Guillette and Gunderson 2001; Toppari 2002). However, there is doubt as to whether PCBs can damage human reproduction, that is, whether PCBs alter the accurate transmission of the human genetic information. A recently accomplished EU-funded project (INUENDO project) has explored the potential association between altered human sperm DNA integrity and the presence of POP in blood. A cross-sectional study was carried out involving 707 adult males from Greenland, Sweden, Poland, and Ukraine, which represents the largest survey ever attempted in molecular epidemiology. Serum levels of 2,2',4,4',5,5'-hexachlorobiphenyl (abbreviated as CB-153, a PCB congener) were determined. In order to evaluate the sperm DNA integrity, the study used a sperm chromatin structure assay. The results of the study show that an increased risk of sperm DNA damage is associated with elevated serum levels of CB-153 across all exposure ranges for the European men involved in the study (Spano et al. 2005); therefore, dietary exposure to PCB might have a negative impact on male reproductive capabilities.

CASE STUDY

According to the results derived by the above-mentioned INUENDO project, European reproduction is threatened by PCBs. As stated in the section on historical emissions of PCBs, worldwide production of PCBs is estimated at ~1.5 million tons (UNEP 1999). It is likely that 30% of the production amount was released to the atmosphere, 4% was destroyed, and about 65% is still in use (e.g. small capacitors), stored or deposited in landfills (Full 2001; Holoubek 2000). The PCB situation differs from country to country; e.g. about 400 tons of PCBs have been illegally disposed into harbors and fjords in Norway (Christian and Janse 2005). Some case studies are given below to facilitate understanding of the PCB situation.

Portugal

PCBs are mainly emitted from older electrical equipment that has still been used until today (Full 2001; Holoubek 2000). There are 1,470 tons of PCBs contained in equipment still in operation and 400 tons in off-duty equipment (Christian and Janse 2005). There are four companies that are licensed for temporary storage of PCB waste, but there are no facilities for final treatment in Portugal (Christian and Janse 2005).

Czech Republic

In the former Czechoslovakia, 21,500 tons of PCBs (plus 1,600 tons of PCB wastes) were produced from 1959 to 1984 (Delor chemical plant in eastern Slovakia); 46% of the total PCB production was exported, and the remainder was supplied to the home market of the former Czechoslovakia (Holoubek 2000). Within both countries (Czech Republic and Slovakia), PCB formulations are still used in the closed systems (e.g. electrical devices such as capacitors and transformers). Currently, waste land-filling is considered to be the most significant source of environmental PCB pollution in these countries. Estimated contribution of applied paint to total PCB pollution within Slovakia is about 5%, and that of industrial and municipal waste incinerators is 10% (Holoubek 2000).

Belgium

A food contamination incident involving PCBs occurred in Belgium in January 1999 (Bernard et al. 1999; Larebeke et al. 2001): a large tank of fat recirculation was contaminated with PCB oil. Refined fat was sold to more than ten animal feed factories, which in turn sold their contaminated feed to farms (mostly chicken farms). The problem was noticed when the chickens showed symptoms of toxicity: low fertility and deformed chicks. The contamination of chicken feed was essentially a PCB problem (not a primary dioxin problem). The highest concentration of PCBs in chicken fat was 51 mg · kg⁻¹ (Σ7PCB in fat), and the dioxin concentration was 0.0026 mg · kg⁻¹ (as I-TEQ in fat) (Bernard et al. 1999). PCBs were later (in summer 1999) found also in pork. In chicken or chicken feed samples, there were 50,000 times more PCBs than dioxin, and the ratio was fairly constant. This means that monitoring in this specific case can be based on PCBs. The cost of PCB analysis is prohibitively expensive (in the order of 1000 Euro per sample) and the analysis is time-consuming (about 1 month) (Kikuchi 2002). Therefore, routine monitoring cannot be based on congener PCB analysis using mass spectrometry.

The Netherlands

There were 22,991 capacitors and 1,222 transformers with PCBs (790 tons in total) in the 1980s, and their owners made use of the opportunity of obtaining a subsidy for disposal and replacement of PCB-containing equipment (Christian and Janse 2005). During 1988 to 1992, almost 2,100 tons of PCB-containing waste was removed by 2,000 companies. Figure 5A illustrates the Dutch scheme for PCB treatment, and Figure 5B shows the treatment realized during 1993 to 2003.

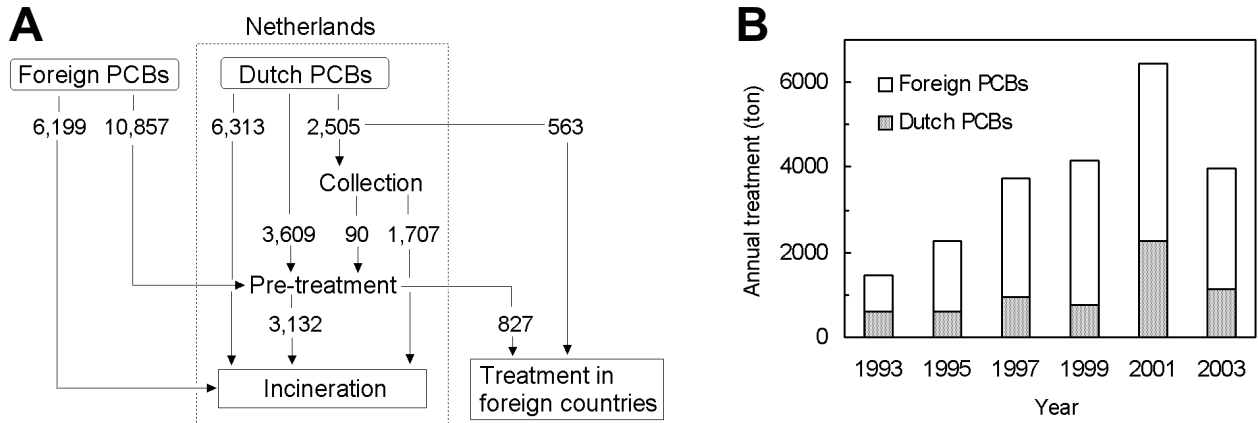


Figure 5. Treatment of PCB waste in the Netherlands (redrawn from Christian and Janse 2005). A – Dutch scheme for PCB treatment and amounts (ton); B – trend of PCB treatment.

It follows from Figure 5A that the Dutch scheme for PCB treatment has flexibility; the Dutch facilities make it possible to treat not only domestic PCBs but also foreign PCBs. Furthermore, Dutch PCB waste is sometimes exported. It can be considered that the Netherlands chooses the most convenient option for PCB treatment at the time. The trend toward PCB treatment (Figure 5B) shows that the Netherlands plays an important role in the European strategy for PCB abatement; a great amount (about 39,000 tons) of PCB waste was treated in the period 1993-2003, and over 40% of the total PCBs were imported.

remaining quantity of PCBs in Europe is 1.1 million tons. The amount of pure PCBs in the liquid is less. Figure 6a shows the remaining quantity of PCB-containing material/equipment in selected countries.

According to article 11 of the European Directive No. 96/59/EC, the plan should be focused on the cleaning and disposal of PCB-containing equipment (both inventoried equipment and uninventoried small equipment). The way most member states handle the removal of possible PCB-containing equipment is to set up a disposal system for such equipment as waste. The duty to dispose of PCB-containing equipment/material means that there must be sufficient capacity for the pre-treatment and disposal of such waste. In Europe, the pre-treatment facilities drain the PCB liquid and then rinse the remaining equipment. The PCBs are incinerated and the metal is suitable for recycling. Figure 6B shows treatment facilities for PCBs in selected countries.

REMAINING PCBs AND TREATMENT CAPACITY

According to a report by the Chemical Legislation Network (Christian and Janse 2005), it is estimated that the total

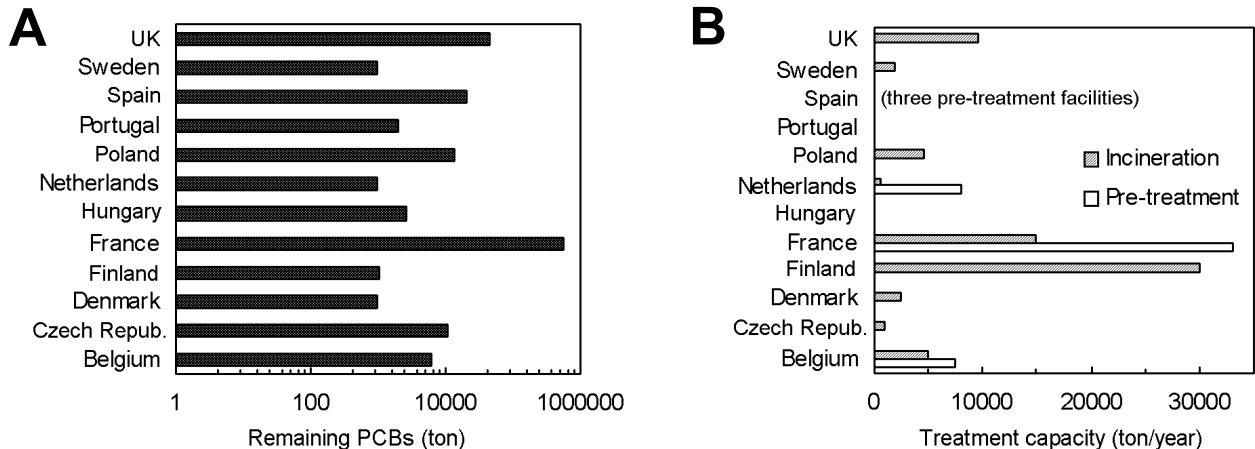


Figure 6. Remaining amount of PCB and treatment facilities in selected countries (redrawn from Christian and Janse 2005). A – remaining amount of PCBs contained in equipment and material (the amount is converted to tons on the basis of gross weight); B – facility capacity (ton · year⁻¹) for PCB treatment (incineration capacity is based on average PCB content in liquid waste). Spain has three pre-treatment facilities, but no PCB incinerator.

The EU 25 member states have about 50,000 tons · year⁻¹ pre-treatment capacity for PCB equipment and about 120,000 tons · year⁻¹ incineration capacity.

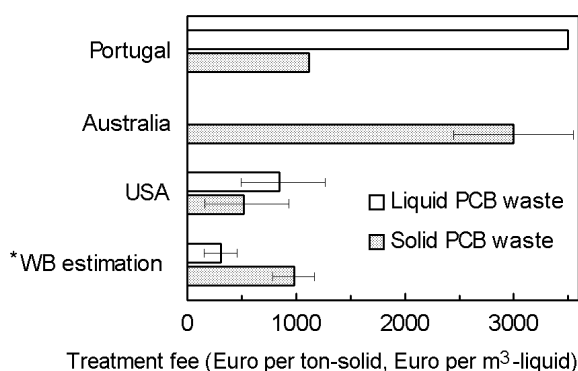
The aim of the council directive (see introduction) is to eliminate PCBs completely. A simple calculation is made to estimate how long it will take to eliminate PCBs completely: based on the remaining amount (1.1 million tons) and the pre-treatment capacity (50,000 tons · year⁻¹), it will take 22 years to pre-treat all PCBs; and based on the remaining amount and the incineration capacity, it will take about 10 years to incinerate all PCBs. This implies that European people (as well as Polar animals, terrestrial and aquatic species) will continue to be threatened by PCBs for a long period of time.

TREATMENT TECHNOLOGY AND STRATEGY

According to a regulation (No. 761. 60 a) of the U.S. Environmental Protection Agency (OECA 2004), PCBs at concentrations of 50 ppm or greater must be disposed of in a high-temperature incinerator: the temperature is maintained at 1,200°C for 2-second residence times or 1,600°C for 1.5-second residence times. That is, incineration is the standard method of PCB destruction because the destruction efficiency can reach 99.9999% and the choice of incineration is often based on cost performance (OPPT 1997).

Cost performance

According to a report by the World Bank (EEC 2004), the capital cost of a high-temperature incinerator is estimated at 30 million to 40 million Euro on the basis of a 6 tons · h⁻¹ kiln type. The European countries shown in Figure 6B also use thermal decomposition at high temperatures. Prices of PCB treatment are compared among different regions in Figure 7.



*World Bank estimation

Figure 7. Comparison of PCB treatment fees by region on the basis of incineration [derived from DEH (1997); EEC (2004); OPPT (1997); Prolixo (2004); SUCH (2001)]. Exchange rate: 1.00 Euro = 1.25 US dollars = 1.67 Australian dollars.

Figure 7 shows that the treatment fees vary widely. The following reasons are considered: (i) *Australia* – at the time the cost estimation was made, incineration of PCB waste was still in the project phase. The presented costs may have referred to New Zealand's export of hazardous wastes (8,950 Euro per m³-liquid PCB) (DEH 1997), so the estimated fee is comparatively expensive; (ii) *USA* – PCB incineration has been introduced and generalized, so the fees are cheaper than those in the other countries; (iii) *World Bank* – it can be considered that the fees refer to the USA's market of PCB treatment, so the reported fees are similar to those in the USA; (iv) *Portugal* – as stated in the section presenting a case study, there are no treatment facilities. It is natural that the costs for handling and transport are more expensive than those in the countries having treatment facilities.

Although the number of data in Figure 7 is statistically insufficient, a simple conclusion is derived: PCB treatment is comparatively cheap in the countries having treatment facilities (e.g. the USA), but it is expensive in the countries without such facilities (e.g. Portugal).

Alternative technologies

High-temperature incineration is now one of the leading methods of PCB treatment as stated above, but various processes for PCB treatment have been developed (Rahuman et al. 2000): Super Critical Oxidation, Electrochemical Oxidation, Solvated Electron Technology, Chemical Reduction Reaction, Molten Metal Pyrolysis, Molten Salt Oxidation, Catalytic Hydrogenation, Solvent Extraction-Chemical Dehalogenation-Radiolytic Degradation, Solar Detoxification-Photochemical Degradation, Thermal Desorption Integrated Technologies (Thermal Desorption-Catalysed Dehalogenation, Thermal Desorption-Pyrolysis, etc.), and Biological Technologies (e.g. Bioslurry). Some typical processes are discussed as follows:

(i) *co-incineration of PCB in cement kiln* – this technology is the generation of energy which uses waste as a regular or additional fuel, and this co-incineration is in common use particularly in the cement industry in Europe (EUFP 2000). The reason for this is that the cement industry relies heavily on an energy-intensive process. About 400 tons of PCBs were treated by waste co-incineration using cement kilns during 1990-1995 in Norway (Christian and Janse 2005). When waste-containing chlorine is burned together with lime-bearing materials for clinker production in a cement kiln, the final product contains a greater amount of leachable chlorine (Kikuchi 2005) and a smaller amount of SiO₂. This poor quality reduces the application range of the obtained cement. Given the correlation between the chlorine content of waste and cement quality, it cannot but be considered that waste co-incineration in a kiln used for cement production has severe operational limitations (Kikuchi 2005).

(ii) *dechlorination* – the most common technology is based on the use of metallic sodium to dechlorinate the PCB molecules and yield an oil which can be re-used, whether in the transformer or for some other use (UNEP Chemicals 2000). The sodium technology uses an amount of reagent which is proportional to the initial PCB content of the oil. In addition, the time of reaction will also be dependent on this initial PCB level. This means that the cost of the decontamination can only be expressed in terms of the oil contamination level. This is not always taken into account in the costs advanced for sodium processes (UNEP Chemicals 2000).

(iii) *plasma-related technology* – the principle is to subject a stream of the material to be treated to a high-energy electrical discharge, and extremely high temperatures can be attained in the plasma arc (UNEP Chemicals 2000). This condition can be used to convert many complex organic compounds into simpler, harmless molecules such as carbon dioxide, water and hydrochloric acid. Although the plasma treatment is technically sophisticated, treatment costs can be relatively high and the technology is not in widespread use. It is reported that the emissions of dioxins, Cd and Pb are heightened by chlorinated waste in plasma-arc melting at a melter temperature of 1,000 to 1,700°C and at an oxidizer temperature of 900 to 1,000°C (Yang and Kim 2004): significant amounts of Cs, Cd and Pb are vaporized during the high-temperature melting. This experiment suggests that wet scrubbing and/or fine filtration cannot effectively remove dioxins/furans (PCDD/Fs) and Hg species from the flue gas (gas phase and fine-particle phase) (Yang and Kim 2004). If this system is put into industrial practice, an appropriate measure should be required to capture dioxins/furans and volatile metals.

(iv) *low-temperature thermal desorption* – this technique has been developed to remediate PCB contaminated soil and not to treat PCBs themselves. After iron nano-particles and the PCB-contaminated soil are mixed in water, the contaminated soil is treated by thermal destruction at 300°C (Vanasi et al. 2007).

CONSIDERATION

A high concentration of PCBs ($>20 \text{ pg} \cdot \text{m}^{-3}$, see Figure 3B) is observed in the European atmosphere, and not only Polar animals but also European male fertility has been affected by PCB risk. According to a recent case study in Switzerland (Morf et al. 2007), the average PCB concentration is $13 \text{ mg} \cdot \text{kg}^{-1}$ in waste electrical and electronic equipment. The average level indicates a notable decrease of PCB application in electrical and electronic equipment, but this level is not null. A European Directive (No. 76/769/EEC) aims to eliminate PCBs completely, and the presented calculation suggests that it will take over 20 years to achieve the directive's goal if things come to the worst.

Currently available capacity of waste incineration

The total amount of waste would be estimated at 1.3 billion tons $\cdot \text{year}^{-1}$ in Europe (ETCW 1999). Compared with this total amount, PCB waste is a small portion; hence, it is not economical to install an incinerator only for PCB treatment.

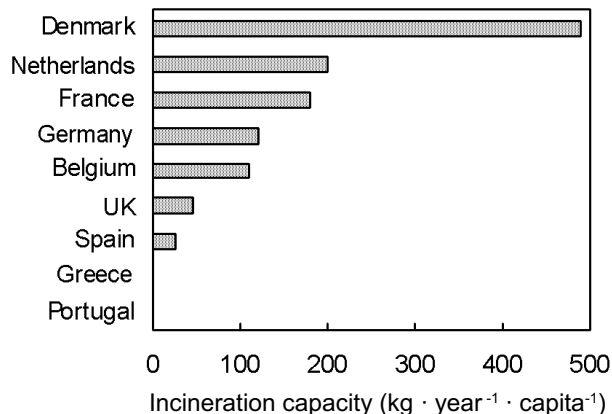


Figure 8. Available capacity for municipal waste incineration in selected EU countries (redrawn from ETCW 1999).

Figure 8 illustrates the available capacity of incineration for municipal waste. There is a high degree of variation in incineration capacity; about 500 incineration plants are reported in operation, and their total capacity is almost equivalent to about 17% of municipal waste (ETCW 1999). This means that landfill is a common mode of municipal waste treatment in most European countries. Historically, the main purpose of waste incineration was to reduce the amount of waste to be landfilled, and usual incineration can reduce municipal waste to about 30% of its original volume (Shigaki 1998). It is assumed that the number of incineration plants for municipal waste will increase since the European waste directive called for a 30% reduction in the amount of waste sent to landfill.

Preferable multi-use incinerator

In a waste incineration project, it seems practical to build an incinerator which can safely burn both municipal waste (e.g. $> 850^\circ\text{C}$ and a waste retention time of 2-3 seconds) and PCBs (e.g. $> 1,200^\circ\text{C}$ and a waste retention time of 2 seconds). As stated, high-temperature incineration has been a mainstay of PCB treatment from the viewpoints of removal efficiency and cost performance. Considering the technical strategy, multi-use incineration is desirable because it will be able to burn municipal waste after the complete elimination of PCBs according to the PCB directive. Wastes are roughly classified into 20 categories, and the list of hazardous wastes contains 236 codes in Europe (EEA 2003). As it is not economical for one incinerator to treat one specific type of waste, the process

used in the project should treat as many types of waste as possible, and it is desirable that the plant constructors commercialize such a system at reduced cost.

CONCLUSIONS

In Europe, renewable energy increased by 15% between 1992 and 1999 (EEC 2004). In Western Europe, this growth was supported by policy interventions. In Central and Eastern Europe (CEE), the most of growth came from an expansion of energy recovery from waste combustion, but this does not seem to be linked to any policy initiatives (EEC 2004); there is therefore a good possibility that political coordination may contribute to spread the multi-use incineration (with energy recovery) in CEE. If so, CEE will have an important part in the European strategy for the virtual elimination of PCBs.

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