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REVIEW ARTICLE

Applications of polychlorinated biphenyls

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Abstract

Background, aim, and scope In the 50 years or so that polychlorinated biphenyls (PCBs) were manufactured in the USA and elsewhere, they were widely used in numerous applications because of their desirable properties. The purpose of this paper is to review and summarize in one place the factual information about the uses of PCBs, as well as to correct some misconceptions that have arisen over the years. The focus is on applications in the USA for which there is ample documentation. However, use patterns were probably similar worldwide.

Materials and methods Review.

Results PCBs were used primarily as electrical insulating fluids in capacitors and transformers and also as hydraulic, heat transfer, and lubricating fluids. PCBs were blended with other chemicals as plasticizers and fire retardants and used in a range of products including caulks, adhesives, plastics, and carbonless copy paper. In the USA, PCBs were manufactured from 1929 through mid-1977, although many products remained in service for decades after the manufacture of PCBs was terminated. This article reviews the historic uses of PCBs in the USA and discusses, where possible, the relative sales volumes. Especially with smaller volume, military, and third-party uses, documenting a use and/or differentiating between a commercial use and an experimental test batch is not possible.

Discussion A major contribution of this paper is to differentiate reported commercial applications of PCBs that can be documented from those which cannot. Undocumented uses may include actual minor uses as well as reported applications that are unlikely ever to have been commercialized.

Keywords PCBs · Polychlorinated biphenyls · Aroclor · Capacitor · Transformer

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1 Introduction

In July of 1977, the sole US manufacturer of commercial polychlorinated biphenyls (PCBs), the former Monsanto Company[1] voluntarily ceased manufacturing the products at their plant in Sauget, IL (production at the Anniston, AL, plant had ceased in 1971). Starting about a decade previously and continuing for the succeeding three-plus decades, PCBs have been among the most studied groups of chemicals. Publications number in the tens of thousands and the publication rate shows no sign of slowing. PCBs remain an economic force over 30 years after the last products were made. Issues relating to PCBs provide professional opportunities, funding, and income to numerous regulators, academic, and government research scientists, consultants, remediation firms, and attorneys.

In the 40 years or so that PCBs have been in the eyes of all these various parties, as well as those of the public itself, much has been learned. However, some of what has been "learned" is based on misunderstandings, apocrypha, and careless repetition of undocumented "facts" that just were

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[1] All succeeding mentions of Monsanto Company refer to the "old" Monsanto Company, now known as Pharmacia. The company now known as Monsanto was chartered in 2000 and is a manufacturer of agricultural products.

not so. Some of these "myths" are merely amusing, for example, the myth that the "12" in the names of Aroclor products stood for the 12 carbon atoms in the biphenyl molecule. Others have had more serious implications as discussed below.

Here, we review a range of factual information about the uses of PCBs, as well as to correct some of the misconceptions that have continued to be expounded over the years. PCB nomenclature, manufacturing, and properties are discussed for perspective. The focus is on applications in the USA for which there is ample documentation. However, used patterns were probably similar worldwide. PCBs synthesized incidentally to other chemistries and incorporated into products as inadvertent constituents are not addressed here. Also, the manufacture and use of single congeners for research and as analytical standards are outside the scope of this article. Finally, issues related to the presence of PCBs in the environment and associated potential exposures are not dealt with here.

2 A brief history

PCBs were first described in the German chemical literature in the 1880s (Schmidt and Schultz 1881). PCBs were first manufactured commercially in 1929 by the Swann Chemical Company in Anniston, AL. Theodore Swann had developed a commercially viable process to manufacture biphenyl from benzene by bubbling benzene through molten lead. Chlorination of the biphenyl was one of many routes explored to develop commercial uses for the biphenyl (Penning 1930). General Electric was among the companies which tested this new product. They were looking for a flame-retardant transformer fluid to use in locations where mineral oil fires put persons or property at risk. General Electric recognized that PCBs were ideal fluids for this application and patented various PCB-containing products in the early 1930s.

In 1935, the Swann Chemical Company, including Swann's Anniston, AL, plant was purchased by the Monsanto Company, based in St. Louis, MO. Swann's line of polychlorinated polyphenyl products, known as Aroclor® products, was among the product lines now manufactured by Monsanto at Anniston.

In the late 1930s, a second manufacturing facility was constructed in Sauget, IL. During World War II, the manufacture of PCBs was taken over by the US Government, because of their essential uses in support of the war effort. After the war, the uses of PCBs expanded into a number of functional areas, including flame retardant heat transfer fluids, hydraulic fluids, and plasticizers. These uses are described in detail below.

The discovery and subsequent investigations of the presence of PCBs in the environment is a tale oft-told and will only be quickly summarized here. The first mention of PCBs in the environment was in the British science news magazine *New Scientist* in December of 1966 (Anonymous 1966), reporting the findings of Sören Jensen and colleagues in Stockholm. The first mention of the Swedish work in the USA was in January 1967 (Anonymous 1967). Over the next several years, additional studies were published, including the December 1968 publication in Nature first reporting PCBs in US birds (Risebrough et al. 1968). In 1972, Jensen (1972) published *The PCB Story*, doubtless thinking it important to commemorate this historic tale before we all moved on to other scientific challenges. As we know, the PCB story was far from over in 1972.

By early 1970, Monsanto had undertaken a program to address the presence of PCBs in the environment. Customers were notified of the developing information about PCBs in the environment, and Monsanto introduced a label warning users to prevent environmental discharges. Monsanto also voluntarily withdrew PCBs from all markets which were considered likely to lead to environmental discharges. Sales were restricted to a limited number of manufacturers of electrical equipment for uses in nominally closed systems, such as capacitors and transformers. Consideration was given to early cessation of manufacture, but a US Government inter-departmental task force noted in May of 1972 that the continued use of PCBs in electrical equipment was essential to the safe delivery of electrical power in the USA (ITF 1972).

In 1968, the Yusho incident occurred in Western Japan, mainly in the Fukuoka and Nagasaki prefectures. "Yusho" is a Japanese word meaning "oil disease"; it is not the name of a geographical location in Japan, as is often stated. The incident did not occur "in Yusho." Thermally degraded Japanese PCB-containing heat transfer oil had leaked into rice oil during processing. The rice oil was subsequently consumed by residents in Western Japan. The details of this incident have been thoroughly covered in the scientific literature (Kunita et al. 1984) and in books (Erickson 1997 and citations therein). The thermal degradation of the fluid had resulted in elevated levels of polychlorinated dibenzofurans and other chemicals in the fluid and subsequently in the rice oil. Although investigations continue to this day, it is widely acknowledged that the primary causative factor of Yusho was the polychlorinated dibenzofurans, since Japanese electrical workers with comparable levels of PCBs in their bodies did not exhibit the symptoms of Yusho.

After Monsanto was notified by its customers that acceptable substitute fluids for PCBs in electrical equipment were available, Monsanto ceased production of PCBs in 1977, 2 years before the EPA's ban on the manufacture of

PCBs was published in May of 1979. However, PCB manufacturing in several European and Asian countries continued well into the 1980s and probably later. Today, the intentional manufacture of PCBs is not known to be occurring anywhere in the world, except for the synthesis of small amounts for research purposes.

2.1 PCB manufacturing process

Monsanto manufactured PCBs by the direct chlorination of biphenyl (Hubbard 1964). Ferric chloride was used as a catalyst. When the desired degree of chlorination was attained, as determined by the specific gravity, the crude liquid Aroclor product was pumped to a tank where residual hydrochloric acid (HCl), which was a byproduct of the chlorination reaction, was blown out with air. Following treatment with lime to neutralize any residual acid, the crude mixtures were refined by vacuum distillation. To prepare electrical grade Aroclor 1200 series products, the distilled material was treated with attapulgus earth (fuller's earth) to remove electrically conductive impurities such as traces of water and HCl and thereafter filtered.

The complex congener composition of the various Aroclor PCB products was determined by the chemistry and physics of the chlorination process. There is a frequent misunderstanding that Monsanto and the other manufacturers somehow manufactured and blended the individual congeners to produce the various products. This is simply not the case. While it is not possible to delineate all of the reaction kinetics, a couple fairly simple considerations may help to illustrate the considerations that determined the congener mixes. Readers who either enjoyed or suffered through college organic chemistry may recall that substituents on benzene rings "direct" further substitutions to either the *ortho*/*para* or the *meta* positions on the rings. The second benzene ring in the biphenyl molecule is an *ortho*/*para* director, so substitution is much more common in those positions than in *meta* positions. Also, the chlorines tend to be distributed somewhat equally between the two rings, so that congeners with three or more chlorines on one ring and none on the other are not present in actual product mixtures, even though such congeners frequently serve as research curiosities. Although there is a consensus on the general homologous compositions of the major Aroclor products (see Table 1), characterization of the composition of the commercial mixtures at the congener-specific level is much more complex and remains a subject of continuing research.

The manufacturing process also helps explain why PCB products of the same chlorination level are remarkably similar among different manufacturers and among batches from the same manufacturer. As long as the processes are well-controlled, the reactions will occur in the same way in every batch for every manufacturer. Of course, there will be minor variations, but the major components will always be major components, and the trace components will always be trace components.

Table 1 Comparison of commercial PCB mixtures

Aroclor	Average No. Cl/Molecule	Approximate Weight% Cl
1221	1.15	21
1232	2	32–33
1242, 1016	3	40–42
1248	4	48
1254	5	52–54
1260	6–6.3	60
1262	6.8	62
1268	8.7	68
1270	10	71

Source: (Brinkman and De Kok 1980)

2.2 The naming of cats: PCBs

As we were reminded in the musical *Cats*, every cat has three names. The same is true of PCBs, although some have even more names. For example, 3,3′,4,4′-tetrachlorobiphenyl is also known as PCB 77, is a non-*ortho* PCB or coplanar PCB, and has CAS Registry Number 32598-13-3. Of course, it is also a congener and an isomer, and it may be a component of a commercial mixture, such as one of the Aroclor products. We sort out this confusing nomenclature here.

The term "congener" has come to be applied to any single member of a class of related compounds, such as PCBs, which are the class of compounds comprising molecules with 1–10 chlorine atoms attached to the two rings of biphenyl. Despite the linguistic inconsistency, monochlorobiphenyls are included in all PCB discussions. Unchlorinated biphenyl is never included as a PCB. There are 209 PCB congeners. These congeners can be further classified according to the number of chlorines attached to the rings. Thus, there are 10 "congener classes," ranging from monochlorobiphenyls (three class members) through pentachlorobiphenyl (46 class members) to decachlorobiphenyl (one class member). When grouped by degrees of chlorination, the congener classes are often referred to as "homologs," although that term is strictly applicable only to groups of chemicals with increasing carbon chain lengths. However, the application of the term to PCBs and other groups of chlorinated compounds is widespread in formal and informal writing and must be considered an accepted use.

The term “isomer” refers to one of a group of chemicals that have the same molecular formula, i.e., they comprise the same elements and the same numbers of those elements. Thus, the 42 members of the congener class of tetrachlorobiphenyls are isomers of one another. They all have the molecular formula $C_{12}H_6Cl_4$. (N.B., there are not 209 *isomers* of PCBs, because PCBs as a group have 10 possible molecular formulae.)

Of course, like every chemical, each PCB congener has a precise chemical name in accordance with the system established by IUPAC. In our example above, that name is 3,3′,4,4′-tetrachlorobiphenyl. That name can only apply to that specific congener, and it uniquely specifies the number and location of the chlorine atoms on the biphenyl rings. That naming system is precise and works well for congeners with only a few chlorine atoms, but it quickly becomes cumbersome as the number of chlorines increases. Accordingly Ballschmiter and Zell (1980; corrected in Ballschmiter et al. 1992) proposed a numbering system in which each congener was arranged in ascending IUPAC hierarchical order from mono- to decachlorobiphenyl and given a number from 1–209 (the BZ number) to facilitate communication of information about individual congeners. Thus, in the BZ system, 2-chlorobiphenyl is PCB 1; 3,3′,4,4′-tetrachlorobiphenyl is PCB 77; and 2,2′,3,3′,4,4′,5,5′,6,6′-decachlorobiphenyl is PCB 209.

All 209 congeners, “PCB,” the ten homologs, Aroclor products, and other PCB-related mixture terms have a unique number assigned by the Chemical Abstracts Service, which has assigned numbers to over 50 million organic and inorganic substances. 3,3′,4,4′-Tetrachlorobiphenyl has CAS Registry Number 32598-13-3. Numbers are assigned when the chemical is reported in the literature, so the CAS numbering system is not sequential. For example, the next congener on the BZ list, 3,3′,4,5-tetrachlorobiphenyl, has a CAS RN of 70362-49-1. A comprehensive list of all congeners with IUPAC, BZ, and CAS numbers can be found in Appendix A in Erickson (1997).

Primarily to facilitate discussions of the toxicological properties of certain PCB congeners, the *ortho*, *meta*, and *para* designations are used to classify PCBs according to their potential ability to bind to the aryl hydrocarbon (Ah) receptor in animal cells. The Ah receptor is a cellular receptor that binds planar organic compounds such as polychlorinated dibenzo-*p*-dioxins and dibenzo-*p*-furans with high affinity, leading to various toxic effects. The most potent ligand is 2,3,7,8-tetrachlorodibenzo-*p*-dioxin or TCDD. In this classification scheme, *ortho*-chlorines are those in the 2, 2′, 6, or 6′ positions, i.e., those adjacent to the carbon–carbon bond in biphenyl. Likewise, *meta*-chlorines are those in the 3, 3′, 5, or 5′ positions, and *para*-chlorines are those in the 4 or 4′ positions. The significance of this scheme is that PCB congeners with at least four chlorines and with no chlorines in the *ortho* positions can assume the planar conformation necessary for binding to the Ah receptor. These congeners (BZ numbers 77, 81, 126, and 169) are thus frequently called coplanar, non-*ortho*, or dioxin-like PCBs (note that these PCBs are not “locked” into the planar conformation, but they can assume that conformation during rotation around the carbon–carbon bond.) PCBs with at least four chlorines in the 3, 3′, 4, 4′, 5, or 5′ positions and a single chlorine in an *ortho* position are denoted mono-*ortho*-PCBs. These eight congeners bind weakly to the Ah receptor. Lastly, the remaining congeners are designated as either di-*ortho*-PCBs and, more generally, as *ortho*-PCBs.

It should be noted that the four non-*ortho* and eight mono-*ortho* PCBs have been assigned TCDD toxicity equivalency factors by the World Health Organization (WHO) and other organizations to reflect the potential relative potencies associated with binding to the Ah receptor, compared to that of TCDD (Van den Berg et al. 2006).

2.3 The naming of cats: Aroclor products[2]

As noted earlier, Monsanto’s trade name for its line of polychlorinated polyphenyl products was Aroclor®. Readers will please note that there is no “h” in Aroclor (the trademark designation is generally omitted throughout this article to be consistent with common usage). Of course, the most widely known of these products were the polychlorinated biphenyls, but the product line also included polychlorinated terphenyls (PCTs), as well as mixtures and blends of PCBs and PCTs. In the broadest of terms, most PCBs were known as liquid Aroclors, while the term solid Aroclors encompassed PCTs and the most highly chlorinated PCBs.

In general, the naming system for Aroclor PCB products is well known. The trade name Aroclor was followed by a four-digit number (Table 1), in which the first two digits were “12,” designating the product as a refined PCB. The second two digits specified the average percentage of chlorine, by weight, in the particular product. Thus, Aroclor 1242 was a polychlorinated biphenyl product containing 42% chlorine by weight. While 42% chlorine by weight is also the approximate composition of trichlorobiphenyls, the product is a complex mixture of congener classes containing from one to six or seven chlorines. It is not “trichlorobiphenyl,” per se (this frequent misconception is compounded by the naming systems of some non-US PCB products, as will be discussed below).

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[2] Unreferenced Aroclor and other Monsanto product information (Section 2.4) is derived from personal knowledge, RGK.

One frequently reads the myth[3] that the “12” in the product name refers to the fact that there are 12 carbon atoms in the biphenyl molecule, which is decidedly not true. In fact, for every product in the Aroclor 1200 series (refined PCBs), there was a corresponding product in a less well-known 1100 series, the crude PCBs. As noted elsewhere, the final step in the manufacture of the 1200-series PCBs was the distillation of the corresponding crude 1100-series material. Thus, Aroclor 1142 was distilled to produce Aroclor 1242. Further, like PCBs, the PCTs were marketed with a four digit specification, in which the last two digits indicated the percentage of chlorine by weight in the product. However, the first two digits were “54.” Thus Aroclor 5460 was chlorinated terphenyl with an average chlorine content of 60%. If the “12=12 carbon atoms” myth were true, the first two digits of the PCT line would have been “18,” since there are 18 carbon atoms in the terphenyl molecule (the crude PCT products had designations in the Aroclor 5000 series).

The one oft-noted exception to the naming system for PCBs is Aroclor 1016. This product was developed and introduced after 1971, when it became clear that PCB congeners containing three to four chlorines or fewer were fairly rapidly biodegradable, while those with five or more were less so. Aroclor 1016 was produced by distilling Aroclor 1242 to remove the more highly chlorinated congeners to make a more biodegradable product. Further, since it was introduced after Monsanto limited sales of PCBs to manufacturers of electrical equipment for use in closed systems, Aroclor 1016 was predominantly used in capacitors, with some limited use in transformers.

The “1016” designation was an outgrowth of Monsanto’s system for keeping track of materials in the research stage of development. Each new research chemical, whether PCB-containing or not, was given a sequential Monsanto Chemical Substance or Sample number (MCS). Thus, MCS 1016 was the designation of the Aroclor 1242 distillation product that was undergoing research to see if it would be a suitable replacement for Aroclor 1242 in electrical equipment. During the product development stage, both Monsanto personnel and customers began to refer to the research material as simply “1016,” just as they referred to the other PCB products simply by their four-digit name. When MCS 1016 was commercialized, it was called Aroclor 1016, because that is what practitioners were already calling it. The name was not an attempt to disguise the fact that it was a PCB product or to suggest that it had only 16% chlorine. Claims to that effect fail to recognize the developmental history of the product.

Finally, there were also a few products containing both PCBs and PCTs, namely Aroclor 2565 and Aroclor 4465 (which was refined from Aroclor 4065). The Aroclor 6000 series of plasticizers was formulated as blends of Aroclor 5460 and Aroclor 1221. These products served as transitional plasticizers between PCB-containing and non-PCB-containing products. In this series, the final two digits indicated the amount of Aroclor 5460 in the product. For example, Aroclor 6050 contained 50% Aroclor 5460 and 50% Aroclor 1221.

2.4 The naming of cats: other Monsanto PCB products

Aroclor was the dominant trade name for Monsanto’s PCB and PCT products. However, other trade names were used for specific applications, sometimes because the Aroclor product was blended with other chemicals.

Therminol® was the trade name for Monsanto’s line of heat transfer fluids. The original fluids were all in the FR series, where the “FR” referred to the flame retardant properties of the fluids. Only the Therminol FR series fluids contained PCBs (EPRI 1999; Therminol 66 was erroneously noted to be a PCB-containing product at p. 3–12). In fact, with the exception of Therminol FR-0 and Therminol FR-1 Lo-Temp, the Therminol FR products were 100% PCBs: FR-1 (Aroclor 1242), FR-2 (Aroclor 1248), and FR-3 (Aroclor 1254). After Monsanto ceased selling PCBs for open application in the early 1970s, they continued to sell heat transfer fluids and continued to use the Therminol trade name. The Therminol trade name is currently used by Solutia Inc. to which the business was spun off in 1997. Of course, no Solutia-manufactured Therminol fluids ever contained PCBs.

The situation with regard to Monsanto’s former line of Pydraul® hydraulic fluids is not so straightforward. Monsanto’s early line of PCB-containing Pydraul fluids were blends of PCBs along with, variously, hydrocarbon oils, phosphate esters, and other chemicals, as well as additives such as rust inhibitors, viscosity modifiers, and colorants. In most cases, each particular Pydraul product was developed for a specific application, often in association with customers. For example, Pydraul AC was developed specifically for use in air compressors. Accordingly, there is no simple way to know or predict the composition of any particular Pydraul fluid.

As was the case with other “open” uses, Monsanto stopped making and marketing PCB-containing Pydraul fluids in the early 1970s. In many cases, however, the company introduced non-PCB-containing fluids with the same name with a suffix indicating that the fluids no longer contained PCBs. In general, Pydraul fluid names with the suffix “B” indicated the fluid was a transitional fluid, often containing PCTs. Fluids with “C” or higher designations

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[3] For example, http://www.epa.gov/epawaste/hazard/tsd/pcbs/pubs/aroclor.htm and http://en.wikipedia.org/wiki/Polychlorinated_biphenyls. Accessed April 2010.

contained neither PCBs nor PCTs. As with the Therminol name, Monsanto continued to develop and market new, non-PCB containing hydraulic fluids under the Pydraul trade name. Monsanto's hydraulic fluid business was sold in 1986. However, the Pydraul trade name continued to be used by subsequent manufacturers.

Monsanto also marketed a line of aircraft hydraulic fluids with the trade name Skydrol(R). Those fluids were based on phosphate esters and never contained PCBs (Hatton 1964). Phosphate ester-based Skydrol fluids continue to be manufactured and marketed by Solutia Inc. (www.skydrol.com)

2.5 The naming of cats: other manufacturers and products

PCBs were manufactured worldwide through at least the 1980s. Monsanto's Aroclor products accounted for nearly all of the US production. Foreign manufacturers sold similar products under trade names such as Kanechlor® (Japan), Clophen® (Germany), Phenoclor® and Pyralene® (France), Fenchlor® (Italy), Sovol (Russia), Chlorfen (Poland), and Delor® (the former Czechoslovakia). In addition, many use-specific PCB-containing products had identifying trade names. Manufacturers other than Monsanto also added numerical "suffixes" to their trade names to specify the average composition of their product (Erickson 1997 Table 2-V). As noted above, Aroclor 1242 was a complex mixture of PCB congeners from many congener classes, but the average percentage of chlorine closely corresponded to that of trichlorobiphenyl. Comparable products from other manufacturers were Clophen A30, Phenoclor DP-3, and Kanechlor 300; in each case, the "3" referred to trichlorobiphenyl, the average number of chlorines on the biphenyl rings in the particular product. Each manufacturer had similar product names for products with average percentage chlorine compositions close to those of tetrachlorobiphenyl, pentachlorobiphenyl and hexachlorobiphenyl. In some cases, these naming schemes have led to the incorrect inference that the products were composed of "purely" the congener class suggested by the number. However, all of these products were complex mixtures of PCB congeners from many congener classes, just like the Monsanto products.

As noted above, Monsanto used the Pydraul® trade name for PCB-containing hydraulic fluids and Therminol FR® for PCB-containing heat transfer fluids. Further, many users had their own trade names for PCB-containing fluids used in their own products. For example, General Electric's trade name for their PCB-containing dielectric fluids was Pyranol®; that of Westinghouse was Inerteen®; and that of Kuhlman was Saf-T-Kuhl®. Many authors have tabulated and further described those products (Erickson 1997, Table 2-VI; USEPA 2010).

3 Physical properties

The physical properties of the various PCB mixtures have been discussed extensively in other publications, so they will only be briefly mentioned here. Table 2 shows the physical properties adapted from the Monsanto (2004) Material Data Safety Sheet.

Table 2 Properties of selected Aroclor products

PROPERTY	1016	1221	1232	1242	1248	1254	1260	1268
Color (APHA)	40	100	100	100	100	100	150	1.5(NPA) molten
Physical state	Mobile oil	Mobile oil	Mobile oil	Mobile oil	Mobile oil	Viscous liquid	Sticky resin	Off-white powder
Stability	Inert	Inert	Inert	Inert	Inert	Inert	Inert	Inert
Density (lb/gal 25°C)	11.40	9.85	10.55	11.50	12.04	12.82	13.50	15.09
Specific gravity at ºC	1.36–1.37 25°	1.18–1.19 25°	1.27–1.28 25°	1.30–1.39 25°	1.40–1.41 65°	1.49–1.50 65°	1.55–1.56 90°	1.80–1.81 25º
Distillation range (°C)	323–356	275–320	290–325	325–366	340–375	365–390	385–420	435–450
Acidity mg KOH/g, maximum	.010	.014	.014	.015	.010	.010	.014	0.05
Fire point (°C)	None to boiling point	176	238	None to boiling point	None to boiling point	None to boiling point	None to boiling point	None to boiling point
Flash point (°C)	170	141–150	152–154	176–180	193–196	None	None	None
Vapor pressure (mm Hg @ 100°F)	NA	NA	0.005	0.001	0.00037	0.00006	NA	NA
Viscosity (Saybolt Univ. Sec.	71–81	38–41	44–51	82–92	185–240	1800–2500	–	–
at 100°F) (centistokes)	13–16	3.6–4.6	5.5–7.7	16–19	42–52	390–540	–	–

NA not available

Individual PCB congeners are white, crystalline materials. However, as shown in Table 2, the various mixtures are liquids (less chlorinated) or resinous (more chlorinated) because of the mutual melting point depression effects of the congeners. As expected, the physical properties among the mixtures vary according to the amount of chlorine in the products. Specific gravity, boiling point, and viscosity increase as the chlorine content increases, while the water solubility and vapor pressure decrease.

As has been often noted, the very properties that made PCBs desirable for numerous industrial applications were those that contributed to the environmental persistence of the more highly chlorinated congeners. PCBs were resistant to chemical and thermal degradation, as well as to biodegradation.

Of course, the most important property of PCBs was their fire resistance or, alternatively, their flame retardant properties. When PCBs were involved in fires, the primary product of combustion was hydrochloric acid, which is not flammable, so the products of combustion served to quench the fire. Thus, PCBs were highly desirable for applications where fire was a threat to life and property, such as in electrical equipment in commercial buildings and hospitals, in hydraulic systems in foundries, and in heat transfer systems.

4 Uses

4.1 General use categories

Commercial PCB mixtures were used in a wide variety of applications, including dielectric fluids in capacitors and transformers, heat transfer fluids, hydraulic fluids, lubricating oils, and as additives in paints, carbonless copy ("NCR") paper, adhesives, sealants, and plastics. By far, the preponderance of the PCBs was used in capacitors and transformers. Their commercial utility was based largely on their chemical stability, including low flammability, and desirable physical properties, including electrical insulating properties. PCB production and use has been thoroughly reviewed (Durfee et al. 1976; EPRI 1999; Erickson 1997, 2001; Johnson et al. 2006; WHO 1993).

As reviewed by the WHO (1993), PCB use can be divided into three categories:

- *Completely closed systems* (electrical equipment such as capacitors and transformers)
- *Nominally closed systems* (hydraulic and heat transfer systems, vacuum pumps)
- *Open-ended applications* (Major: plasticizer in PVC, neoprene, and other chlorinated rubbers. Other: surface coatings, paints, inks, adhesives, pesticide extenders, and microencapsulation of dyes for carbonless copy paper. Also: immersion oils for microscopes, catalysts in the chemical industry, casting waxes (decaCB), cutting oils, and lubricating oils)

These use categories had different implications for the introduction of PCBs into the environment. Some uses, like carbonless copy paper, resulted in environmental discharges through the recycling of the paper. Other uses, such as caulks, were intended to remain in place for extended periods. The majority of the PCBs were sealed in electrical equipment, where the only environmental impact would have been from accidents, maintenance, or disposal after the original PCB-containing materials had remained in service for years or even decades.

With increased interest in the environmental impact of PCBs, the sale of PCBs for so-called "open" uses, which could lead to near-term release into the environment if not managed properly, were voluntarily curtailed by Monsanto. By 1972, Monsanto had restricted PCB sales to electrical equipment applications.

Durfee et al. (1976) prepared a 489-page report, "PCBs in the United States—Industrial Use and Environmental Distributions," that was published by EPA. This report is cited frequently in this article and a famous table on the "End-Uses of PCTs and PCBs by Type" has been extensively referenced (ATSDR 2000; Johnson et al. 2006; WHO 1978). Durfee's end-use table summarized the report's text and provided a good synopsis of mid-1970s public information on PCB use. Since that time, additional documentation and additional perspectives allow us to improve upon Durfee's classic work, as discussed in this paper.

A Subpanel on PCBs under an Ad Hoc Committee on Environmental Health Research under the apparent auspices of the National Institute of Environmental Health Sciences reviewed the environmental impact of PCBs in 1972 (Hammond et al. 1972). This list of uses was important, given the 1972 publication date and the communications with Monsanto officials for other use data.

Durfee et al. (1976) also tabulated the US PCB production and sales as adapted in Table 3 and Fig. 1. Other publications documented similar use patterns in Japan (Hammond et al. 1972), six European countries (Brinkman and De Kok 1980), and in 23 Organization for Economic Cooperation and Development countries as well as the USA (WHO 1978). In aggregate, the foreign manufacturers accounted for nearly 50% of worldwide production (Bletchly 1983). In all cases, capacitor use dominated, followed by transformers, and then the other applications.

Clearly capacitor and transformer fluids dominated the sales with a combined 75% of US sales. We discuss these acknowledged and major uses of PCBs in this article, but we also delve into other uses that may have comprised smaller amounts, unknown to Monsanto or EPA at the time

Table 3 Estimates of cumulative US production and usage over the period 1930–1975 in metric tons (g×10E6)

	Commercial production	Commercial sales	Industrial purchases	% of Production	% of Domestic sales
US PCB Production	635,000				
US Imports	1,360			0.2	
US Domestic Usage		538,000			
Total US Exports		68,000		11	
Use Category					
Petroleum Additive			450	0.07	0.08
Heat Transfer			9,100	1	2
Misc Industrial			12,000	2	2
Carbonless Copy Paper			20,000	3	4
Hydraulics and Lubricants			36,000	6	6
Other Plasticizer Uses			52,000	8	9
Capacitor			286,000	45	50
Transformers			152,000	24	27
Total	636,000	636,000			

Adapted from Table 1.2-1, p. 7 in Durfee et al. (1976) by conversion from pounds to metric tons and calculation of percentages

of Durfee's tabulation. We also discuss undocumented uses. Table 4 presents an overview of commercial uses; Table 5 lists published PCB applications with no known commercial use; the sections that follow provide additional detail.

4.2 Electrical equipment

The vast majority of PCBs were used in capacitors and transformers and other electrical equipment as dielectric fluids. PCBs were used in electrical equipment because of performance and safety attributes. For example, one of the most important factors was their fire resistance or flame retardancy. The Underwriters Laboratories flammability rating for Aroclor 1242 was 2–3, while that for mineral oil was 10–20, compared to gasoline, with a flammability rating of 90–100 (ITF 1972).

4.2.1 Capacitors

The properties of the dielectric liquid impregnating the cellulosic paper are: non-flammability, dielectric constant matching that of paper, low dissipation factor, high dielectric strength, high chemical stability, low vapor pressure, inert decomposition products in an electric arc, low toxicity of the material, and its decomposition products and low cost (ITF 1972).

PCBs fit those criteria. The industry term for this PCB dielectric fluid was capacitor askarel.[4] The capacitor askarels include neat Aroclors 1221, 1242, 1254, and 1016, as well as a mixture of 75% Aroclor 1254 and 25% trichlorobenzene. The ASTM (1991a) has published standard specifications for capacitor askarels. As with transformers, General Electric used the trade name Pyranol and Westinghouse used the trade name Inerteen; both had code numbers to designate the specific type of askarel (Erickson 1997).

Small capacitors contained as little as 2 mL and large capacitors contained up to 27 L PCB (ITF 1972). From 1957–1971, capacitors accounted for most of the PCB use in the USA (Durfee et al. 1976). The start date of 1957 is based on availability of Monsanto records and the statement may apply to earlier years as well. In 1968, 95% of all US production of capacitor liquids was PCBs (ITF 1972). In 1976, 90–95% of all impregnated capacitors manufactured in the USA were of the PCB type (Durfee et al. 1976). In 1979, EPA estimated that "9.56 million pieces of equipment...contain PCB capacitors" (Westin and Woodcock 1979). Unlike transformer askarels, capacitor askarels were generally pure PCB.

Two important types of capacitors were phase-correcting capacitors for power lines and fluorescent light ballasts. In capacitor manufacture, the PCBs were used to impregnate the paper dielectric and fill air voids. Other applications included a wide variety of uses of small capacitors in appliances and other products, such as air conditioner pump motors, submersible water pumps, automobiles, televisions, light fixtures, clothes washers, clothes driers, refrigerators, freezers, and microwave ovens (EPRI 1999). The fluorescent light ballasts contained a PCB capacitor and/or petroleum-asphalt insulating material ("potting") contaminated with PCBs (USCFR 1999).

4.2.2 Transformers

Most power transformers use a liquid to electrically insulate and remove heat from the core and windings. The desired

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[4] Note that "askarel" is not a trade name and is *not* capitalized.

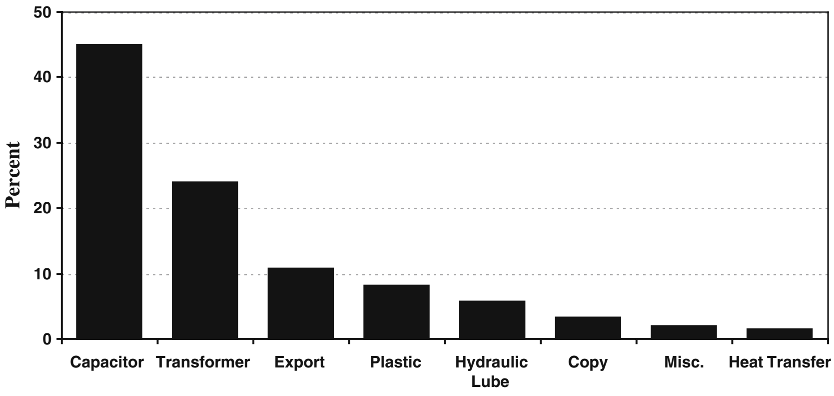

Fig. 1 Applications in the USA based on sales records 1930–1975 (Erickson 2001)

properties are: non-flammability, high dielectric strength, low viscosity, high chemical stability, compatibility with other materials, inert decomposition products, low toxicity of the liquid and its decomposition products and low cost (ITF 1972). PCBs fit those criteria, except that if the appropriate Aroclor fluid was too viscous, it was blended with trichlorobenzene[5] to achieve the desired viscosity. The industry term for this PCB-containing dielectric fluid was transformer askarel. The most common transformer askarels were 60% Aroclor 1260/40% trichlorobenzene (Type A) and 70% Aroclor 1254/30% trichlorobenzene (Type D). The transformer askarels contain other minor components used as free radical scavengers. The ASTM (1991b) has published standard specifications for transformer askarels. General Electric used the trade name Pyranol and Westinghouse used the trade name Inerteen, both with code numbers to designate the type of askarel (Erickson 1997).

Only about 5–10% of transformers were ever manufactured with PCBs during the period when PCBs were used in this application (Durfee et al. 1976; ITF 1972). The vast majority (96% in 1968) used mineral oil for the dielectric fluid, because an askarel transformer cost 1.3 times as much as a mineral oil transformer. The cost of askarel was cited as $1.80/gal and the cost of mineral oil was $0.25/gal, a 7-fold premium for the fluid. (ITF 1972). Fire underwriters required the use of non-flammable dielectric fluids in indoor transformers unless the transformers were in a fireproof concrete vault (ITF 1972).

Askarel dielectric fluids were used in network, pad-mounted, pole-mounted, and precipitator power supply transformers containing 200–2,000 kg in each unit with an average of about 1,400 kg (ITF 1972). Specifically (Durfee et al. 1976):

- "Furnace transformers used in the hot, dirty atmosphere in proximity to glass-melting and induction furnaces... contain 900–1800 kg of askarel each..."
- "Rectifier transformers used for large rolling mills and DC [direct current] industrial power supplies... contain about 8600 kg of askarel..."
- "Railroad transformers used on-board in electric locomotives or multiple unit electric railroad cars...contain 300–1100 kg of askarel in each unit... since a tunnel fire in 1940 caused by an oil filled locomotive transformer, Penn Central will not allow any oil containing transformer equipped locomotive into New York City."
- Reactors: "During power surges they choke the voltage and deliver the normal output."
- Grounding Transformers

In large transformers, hundreds of liters of PCB fluid provided insulation between the high voltage core and the tank, which would be grounded.

4.3 Air and gas compressor lubricants

The use of PCBs in air compressor lubricants was driven primarily by two considerations; reduced fire and explosion hazard and lower maintenance costs due to the reduction of carbon deposits on air compressor valves. The first consideration was particularly attractive to customers operating natural gas pipelines and in other operations where high ambient temperatures made the introduction of mineral oil-based lubricants especially dangerous. PCB uses included "gas-transmission turbines, Aroclors 1221 and 1242" (Hammond et al. 1972).

EPA published a synopsis of the use of PCBs in natural gas pipelines, quoted in part here (USEPA 2004).

> Major interstate natural gas pipelines transport natural gas from production areas on the Gulf Coast and western US to local distribution companies that distribute the natural gas to industrial and urban customers. PCBs were used in turbine and air compressors as a hydraulic/lubricant and a plug valve sealant. As part of the normal operation of large turbine compressors, PCB compressor lubricants could leak or blow by pressure seals and enter the

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[5] The term "trichlorobenzene" is used generically herein for various combinations of tri- and tetrachlorobenzenes used in askarel fluids.

Table 4 Commercial PCB uses

Application	Aroclor(s)	Metric Tons ($g \times 10^6$)	Reference[a]
Electrical equipment			
Capacitors (large, small, fluorescent light ballasts)	1242, 1016, (1254)[b]	286,000	Durfee et al. 1976
Transformers	1254, 1260 (1242, 1016)	152,000	Durfee et al. 1976
Transformer equipment			EPRI 1999
- slip gears			EPRI 1999
- phase converters			EPRI 1999
Slip motors			EPRI 1999
Electromagnets			EPRI 1999
Hydraulics/Lubricants/Heat Transfer Fluids			
Air Compressor/Gas Transmission Turbine Lubricants	Pydraul	G, A	Hammond et al. 1972; USEPA 2004
	Turbinol		
	Santovac		
	1221, 1242		
Heat Transfer	1242, 1248, 1254	9100	Durfee et al. 1976
Hydraulic Fluids (and other lubricants)	1232–1260	36,000	Durfee et al. 1976; Hammond et al. 1972
Vacuum Pumps			EPRI 1999
Motor coolants (mining equipment)	French import		Durfee et al. 1976
Heat transfer systems	1242	9,100	Durfee et al. 1976; Hammond et al. 1972
Vacuum pumps	1248, 1254	A	Hammond et al. 1972
Vapor diffusion pumps			EPRI 1999
Immersion oils for microscopes	1260 & PCT		McCrone 1985
Optical oils in telescopes			EPRI 1999
Cutting oils	1254	A	Hammond et al. 1972
PCBs Incorporated into Products and Materials			
Miscellaneous Industrial		12,000	Durfee et al. 1976
Plasticizers		52,000	Durfee et al. 1976
Carbonless Copy Paper (microencapsulation of ink)	1242	20,000	Durfee et al. 1976; Hammond et al. 1972
Inks	1254		Hammond et al. 1972
Thermographic and xerographic copying		P	ITF 1972
Paints, varnishes, lacquers, and other surface coatings	Many	No info	ITF 1972
Flooring and floor wax/sealants		G,A	USCFR 1999
Coal-tar enamel coatings			USCFR 1999
Pipeline Valve Grease	1268	G	USEPA 2004
Adhesives	1221–1254	P	Hammond et al. 1972; EPRI 1999
Adhesive Tape			USCFR 1999
Caulk and Joint sealants	1254 & other		Multiple (see text)
Gasket sealers			Power Res Inst 1999
Insulation and other building materials	1254, 1268		Multiple (see text)
Rubber products	1232–1254, 1268	A	Hammond et al. 1972; EPRI 1999
Wire and cable coatings	1254, 1260	A, G	Cleghorn et al. 1990; EPRI 1999; USCFR 1999
Die or investment castings	DecaCB (Imported)	13–22/year	Durfee et al. 1976
Petroleum Additive		450	Durfee et al. 1976

[a] In general, we have cited the oldest primary reference for uses, assuming that newer references generally used the first as a source

[b] Aroclors in parentheses are known minor uses

P patent literature, *A* article in published journal, *G* US Government Publication, *M* Monsanto Marketing Literature

Table 5 Published PCB applications with no known commercial use

Application	Aroclor(s)	Metric Tons ($g \times 10^6$)	Reference
Insecticide & bactericide		P, G	ITF 1972
Pesticide extenders	1254	P, A	Hammond et al. 1972
Wax extenders	1242, 1254, 1268	M, A	Hammond et al. 1972; EPRI 1999; Hubbard 1964
Textiles and textile coatings		P	EPRI 1999
Synthetic Resins		A	EPRI 1999
Vinyl chloride polymer films		A	EPRI 1999
Dedusting agents	1254, 1268	A	Hammond et al. 1972
Catalyst carrier		P	ITF 1972

P patent literature, *A* article in published journal, *G* US Government Publication, *M* Monsanto Marketing Literature

> transmission pipeline. These PCBs would generally mix with the "pipeline liquids" already in the transmission lines. The main components of pipeline liquids are water and heavier hydrocarbons that condense-out ("condensate") of the natural gas as pressure drops along the pipeline...
>
> Between 1950 and the early 1970s, Monsanto manufactured and sold several brands of hydraulic/lubricant oils containing PCBs. These included Turbinol 153 that contained 6.4% Aroclor 1221 and 81.5% Aroclor 1242...

4.4 Heat transfer systems

Heat transfer fluids absorb thermal energy from a hot source to provide cooling or to deliver heat. PCBs were used in high-temperature heat transfer systems where their thermal stability, chemical stability and low flammability were needed (ITF 1972). "Flammable heat transfer fluids present a fire hazard if they leak onto a furnace or onto hot surfaces. The use of PCBs prevents this danger" (ITF 1972). Heat transfer systems in petroleum refineries and chemical plants used PCB fluids such as Monsanto's Therminol FR-series heat transfer fluids prior to Monsanto's conversion to non-PCB-containing Therminol fluids.

4.5 Hydraulic fluids

Hydraulic fluids are used as force transmitters. Requirements for such fluids include high lubricity, stability, appropriate viscosity, and compatibility with rubber seals, good fire resistance, and other attributes (ITF 1972). Hydraulic systems are considered nominally closed systems.

In harsh environments in which fire retardancy was particularly valued, PCBs were used as hydraulic fluids (EPRI 1999). Subsurface mining, automobile manufacture, metal finishing, and aluminum industries are examples in which PCB-containing fluids were used. PCBs also served as lubricating additives to hydraulic fluids in extreme pressure applications and as pour-point depressants in hydraulic fluids (ITF 1972). The use of PCBs in hydraulic systems peaked in 1970 when it constituted 15% of the domestic Monsanto sales of Aroclor fluids (Durfee et al. 1976). A US Government panel (Hammond et al. 1972) cited Aroclors 1242, 1248, 1254, and 1260 as having been used in hydraulic fluids and lubricants.

4.6 Vacuum pumps

PCBs were used as diffusion pump oil because of their differential vapor pressure, chemical inertness and other attributes (ITF 1972). Monsanto marketed Santovac 1 and 2 containing 100% Aroclor 1248 and 1254, respectively, for vacuum pump applications.

4.7 Coolants

PCBs were used as engine coolants in mining machinery where fire retardancy was particularly valued. Joy Manufacturing (Pittsburgh, PA) manufactured mining equipment containing motors using PCBs imported from France. Note that this use as a "motor oil" should never be interpreted to include automotive motor oils; there is no evidence of automotive use.

4.8 Microscopy

Aroclors 5442 (a polychlorinated terphenyl) and 1260 were favored by microscopists as mounting media, as components of refractive index liquids, and as immersion oils (McCrone 1985). As recently as 2007, EPA has granted exemptions to "process and distribute in commerce PCBs for use as a mounting medium in microscopy, an immersion oil in low fluorescence microscopy and an optical liquid" (USCFR 2007).

4.9 PCBs incorporated into products and materials

Although PCBs were primarily used as fire-resistant safety fluids for electrical equipment and other applications, over the years they were used as ingredients in products for a variety of additional applications, including the general category of applications known as "plasticizer" applications. As environmental concerns over PCBs began to emerge in the late 1960s and early 1970s, Monsanto voluntarily terminated sales of PCBs for plasticizer applications effective August 31, 1970. Although plasticizer manufacturers could have legally manufactured PCB-containing products until July of 1979, when the Toxic Substances Control Act (TSCA) regulations restricting the use of PCBs became effective, it is not likely that PCB-containing plasticized products were manufactured in the USA after the early 1970s.

4.9.1 Plasticizers

PCBs fell in a broad class of additives called plasticizers that increase flexibility and durability of polymers, plastics, and coatings (Cadogan and Howick 2004; Broadhurst 1972; Hubbard 1964). PCBs mixed well with other components to form a homogeneous composition and had other desirable plasticizer properties (ITF 1972). They were used as plasticizers in paints and coatings where chemical resistance was required (Martens 1968). Other coating performance considerations—air permeability, water permeability, surface hardness—all contributed to the choice of plasticizer.

The PCBs were added in a useful range—too low and they were ineffectual, too high and they imparted undesirable properties to the paint. "If underplasticized, the film will be harder but more brittle and its adhesion may be low. If overplasticized, the film will be softer and more thermoplastic, and consequently will suffer more dirt retention. The permeability of the film is also affected" (Davies 1968). "Aroclor 1221 greatly improves flexibility [to epoxy resins]...Aroclors are especially effective as secondary plasticizers or extenders for polyvinyl chloride. Aroclor 1262, used 1:3 with dioctylphthalate, sharply reduces migration to nitrocellulose lacquers. All Aroclor compounds can be used to improve the chemical resistance of vinyl chloride-vinyl acetate coating formulations" (Monsanto advertisement, *Plastics Technology*, December, 1960).

4.9.2 Carbonless copy paper

Carbonless copy paper was commonly known as NCR paper, variously spelled out as "no carbon required" or "National Cash Register" (a major vendor). "...Aroclor 1242 was used as a solvent for dyes which were micro-encapsulated into microspheres 10–20 microns in diameter and applied to one side of the paper during the coating process" (Durfee et al. 1976). Durfee calculated the average weight percent of Aroclor 1242 in carbonless paper was 3.4%. The US Food and Drug Administration noted that carbonless copy paper contains 3–5% PCBs (38 Fed. Reg. 18101).

Paper recycling or secondary fiber recovery converts waste paper into pulp for new paper products. Because of PCB use in NCR paper and possibly other uses, the recycling processes in numerous paper mills diluted the ~3% PCB content in small volumes of NCR paper through much larger volumes of paper to yield trace concentrations in a variety of media. "Past usage of PCBs in paper coatings and adhesives appears likely, although the quantities used could not have been near the magnitude of PCB in the carbonless copy paper" (Durfee et al. 1976).

4.9.3 Printing

PCBs were added to formulations for several applications in printing:

- Pressure sensitive record paper
- Colored copying paper
- Thermographic duplication paper
- Xerographic transfer process paper

PCBs were added to solvent-free printing mixtures for polyolefin surfaces and in plastic printing plates (ITF 1972).

4.9.4 Paints and surface coatings

The use of PCBs in paints was a plasticizer application. PCBs were a component of specialty paints and coatings to improve performance of the paint in industrial and/or military applications, but they were not for residential or interior decorative use. This application fell within the "open-ended applications" discussed above. The PCBs used for plasticizer applications, including those used in paints, were often sold to independent distributors who resold them to the manufacturers of the ultimate product, for which adhesion, chemical resistance, and/or flame resistance were deemed important. Therefore, product names and PCB composition are largely undocumented. Fabulon floor finish contained PCBs in 1957 (Rudel et al. 2008).

PCBs and other plasticizers were added to coatings in prescribed amounts—generally in the 5–20% range (Chittick and Kirkpatrick 1941; Davies 1968; Parker 1967). EPA has noted (USCFR 1999) that during the 1950–1960 time frame, PCBs were added to paint formulations as drying oils (resins) and plasticizers or softening agents (liquids) in concentrations that range from 10–30% PCBs.

PCBs were a component of an epoxy lacquer used to coat polyethylene and other plastic bottles to make them pliant, impervious, and resistant to aromas, acids, and alkalis. PCBs were used as plasticizers in polyorganosiloxanes that were employed in electrical coatings, insulating tapes, and protective lacquers. PCB-plasticized epoxy resin coatings were used in electrical capacitors, ferrite computer magnet cores, resistors, pipes, blocks, and other surfaces (ITF 1972).

Military and other government uses are not well documented; for example, one source noted PCBs in "wiring insulation, paint, gaskets, caulking, plastic and other non-metallic materials in nearly all of over 100 naval vessels sampled and in service prior to 1977" (Lukens and Selberg 2004). The PCB surface and air concentrations were measured on US Navy surface and submarine vessels to estimate possible exposure of crew members and shipyard workers. Aroclors 1242, 1248, 1254, 1260, and 1268 were found. PCB maximum concentrations of 1–7% were measured in felt insulation, paint, rigid foam, cork, rubber, Armaflex, and Arobol (Still et al. 2003). Military, marine and other applications included waterproofing compounds, anti-fouling compounds, and fire-retardant coatings (USCFR 1999).

"Some older Army, municipal and other water supply systems" used PCB-containing "coal-tar enamel coatings for steel water pipe and underground storage tanks (i.e., AWWA C203 coal tar enamel)" (USCFR 1999). Chlorinated rubber coatings with up to 40% Aroclor 1254 were used as metal coatings where resistance to acids, alkalis, oxidation, electrical conductivity, and properties were important. (Davies 1968; Parker 1967).

"Cumar,[6]" a coating used from 1941–70 to ensure proper curing of concrete used in building 5000–6000 grain silos on farms in the Eastern half of the US, contained ~19% Aroclor 1254 and ~5.4% Aroclor 5460 (PCTs). Upon application and evaporation of the carrier solvents, the PCB content rose to ~32.6%. In some cases, the coatings were eroded by the organic acids produced in the fermentation of the silage, leading to contamination of the silage (Willett and Hess 1975; Willett et al. 1985).

4.9.5 Valve grease and sealant

Aroclor 1268 was used in high-pressure gas pipeline valve grease as a ~10% constituent of the grease. "Rockwell made a plug valve sealant (No 860 and 991) that contained PCB Aroclor 1268 sometime prior to the mid-1970s (Woodyard et al. 1993). The PCB sealant or grease was apparently dissolved by transmission pipeline condensate and spread to other downstream locations" (USEPA 2004, Appendix G).

4.9.6 Adhesives

Because there are myriad surfaces to be bonded with a broad range of functions from temporary to permanent, the world of adhesives is quite large. "Almost every thermoplastic resin is used individually or in resin blends as a hot-melt adhesive. This necessitates a wide range of plasticizers [including] the more resinous chlorinated polyphenyls (higher PCB Aroclors and PCT Aroclors)..." (ITF 1972) Patents were issued for the use of PCBs in:

- Laminating adhesive formulations involving polyurethanes and polycarbonates to prepare safety and acoustical glasses.
- Polyarylene sulfides to laminate ceramics and metals
- Ethylene-propylene copolymer blended with PCB has been used in a hot melt adhesive having improved toughness and resistance to oxidative and thermal degradation...
- Washable Wall Coverings and upholstering materials, made from films of polyvinyl chloride, are claimed to be improved by the addition of PCB to the adhesive formulation.
- PCBs can also be applied in the preparation of polyvinyl alcohol adhesive compositions which are used in the manufacture of envelopes, in self-adhering films, and in the preparation of coatings of pressure-rupturable capsules for adhesive tape. (ITF 1972; The text contains citations to patent literature which were removed for clarity).

Cambric tape containing up to 11% Aroclor 1254 or up to 6% Aroclor 1260 was used as a component of high-voltage electrical cables (Cleghorn et al. 1990).

The bulk of the references to the use of PCBs as adhesives are from patents; there is no evidence how many products were ever in commerce or what PCB volumes they represented.

4.9.7 Caulk and joint sealants

PCBs were used in caulks and joint sealants to plasticize the sealant to maintain a flexible seal between two materials to keep out water, moisture, dust, air, sound, and heat/cold. In some cases, PCBs were incorporated into sealants explicitly to improve fire retardancy (ITF 1972). Polymeric putties were plasticized with PCBs and found to be non-hardening, resistant to moisture and frost and show good

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[6] Cumar is a trade name for "Coumarone-indene resin. Can be used in adhesives. Exhibits good resistance to alkalis, dilute acids, and moisture." http://www.specialchem4adhesives.com/tds/Cumar-LX-509/Neville/529/index.aspx; http://www.nevchem.com/index.asp?pid=02_00_01&pcat=70&prodID=4050 (websites accessed April 2010). There appear to be multiple formulations and there is no implication here that current Cumar formulations contain PCBs.

weather ability. “Elastic pavement or concrete sealing compositions, used for traffic markings, were prepared from coal-tar-polysulfide mixtures which are plasticized with PCB” (ITF 1972). PCB sealants were used in American (Herrick et al. 2004) and European buildings (Andersson et al. 2004; Balfanz et al. 1993; Benthe et al. 1992; Corner et al. 2002; Coghlan et al. 2002; Fengler 1993; Mengon and Schlatter 1993; Priha et al. 2005) and concrete joints and liners in water reservoirs in the USA (Sykes and Coate 1995).

4.9.8 Insulation and other building materials

PCBs were used in fireproof fiberboards and also panels made from starch which can be used for doors, floors, ceilings, and partitions. However, rigid polyurethane foams and hardboard compositions did not show significant increase in flame retardance upon addition of PCBs (ITF 1972). Armstrong manufactured and sold Travertone Sanserra, Santaglio, and Embossed Design ceiling tiles with 4–12% Aroclor 1254 in the coating in 1969–1970 (MMWR 1987). “Wool felt and foam rubber insulation as well as sound-dampening materials have been discovered in naval vessels and may include ships of all types, as well as nuclear submarine reactor compartments” at concentrations up to 70% (USCFR 1999).

Aroclor 1268 was used in various building materials as a fire retardant, including roofing and siding material known as Galbestos. “The main PCB compound used in Galbestos was Aroclor 1268. This construction material was... manufactured from the 1950s to the 1970s by the H. H. Robertson Company” (Panero et al. 2005; USCFR 1999).

PCBs have been found in electric cable components up to 28%, including plastics, foam rubber, rubber, adhesive tape and insulation. These cables were used in marine and industrial applications (USCFR 1999).

4.9.9 Investment casting

“The investment casting [also termed “lost-wax casting”] industry produces precision-cast metal parts and shapes for the aircraft and other machinery manufacturing industries. Approximately 25 of the 135 investment casting foundries in the USA currently use PCB-filled waxes in the manufacture of metal castings. The PCB incorporated in the waxes was decachlorobipheny1 (Fenclor DK or “deka”), which was imported from Caffaro S.P.A., Italy. The remaining foundries use either PCT-filled waxes or unfilled waxes” (Durfee et al. 1976).

4.10 PCB applications with no known commercial use

Monsanto manufactured PCBs from 1935–1977, while foreign manufacturers continued for years after. Aroclor fluids and other trade-named products were industrial products. Although some applications were mandated by industrial codes, building codes, military specifications, and other requirements, most were subject to free-market rules: PCBs were sold and used where the perceived cost-benefit ratio outweighed that of competing chemicals. Prior to the discovery of their environmental persistence, PCBs were specialty chemicals offered for sale, and the manufacturers and customers assertively investigated new applications and marketing.

4.10.1 Examples of patented applications

In 1972, some of the more interesting and non-conventional uses are as follows:

1. Catalyst carrier for polymerization of olefins.
2. Conversion of water-permeable soil to a non-permeable state. Soil is made non-permeable by applying to the soil a composition consisting of an ethoxylene-based resin, polyamide, camphor, and PCB as plasticizer. The composition has a density greater than water, and it hardens under water. It can be applied to river banks, where it flows down the bank, and after hardening, prevents penetration of water (soil erosion-retardant).
3. Combined insecticide and bactericide formulations. The composition contains aldrin or dieldrin, naphthalene hydrocarbons, malathion, methoxychlor, lindane, chlordane, terpineol, and chlorinated biphenyl as active agents.
4. Inhibitors of microbial growth in enamel clay formulations.
5. Plastic sound insulating materials for railway cars.
6. Plastic (PVC) decorative articles which give the impression of internal scintillation.
7. Increasing the density of carbon plates by impregnation with PCB.
8. Graphite electrodes with low thermal expansion coefficients and high bending strengths.
9. Increasing the coke yield from coal pitch. The coke is very hard, dark, and brilliant.
10. As a metal quencher or tempering agent for steel, alloys, and glass.
11. As an aid to fusion cutting of stacked metallic plates without adherence. The cutting is done with an electric arc or oxy-gas torch (ITF 1972; the text contains citations to patent literature which were removed for clarity).

The original citations in this government report are drawn from international patent literature. There is no indication that any of these “uses” ever saw commercial application.

4.10.2 Pesticide extenders

Some chlorobiphenyls were shown to have insecticidal and fungistatic activity; however, they were apparently never used as pesticides although recommended for incorporation into pesticide formulations.

"PCBs are also reported to increase the insecticidal properties of DDT, lindane, organophosphorous compounds, and carbaryl" (Hutzinger et al. 1974).

Although such uses may have occurred in limited situations, at least one attempt to determine whether that was the case was unsuccessful (Reynolds 1971). In an abundance of caution, however, the USDA canceled all registrations of pesticides containing PCBs in 1970 (USDA 1970).

4.10.3 Textiles

PCBs were reportedly used in various textile coatings. Most of the cited uses are in patents and there is no evidence that any products were ever in commercial applications:

- Ironing board covers—PCBs, cellulose acetobutyrate, and aluminum metal particles mixed.
- Delustering rayon
- Coating polypropylene films with mixture of PCBs, UV light absorbers, and antioxidants stabilize against oxidation by sunlight and weathering.
- Polyimide (nylon-type) yarns were flame proofed when treated with PCBs.
- PCBs were a component of a sealing formulation to waterproof canvas.
- PCB additives retarded flame in polyolefin yarns (ITF 1972; The text contains citations to patent literature which were removed for clarity).

4.10.4 Wax extenders

Aroclors 1242, 1254, and 1268 were used as wax extenders (Durfee et al. 1976; Hutzinger et al. 1974). "Carnauba wax may be extended by blending with chlorinated biphenyl in combination with ceresin and paraffin" (Hubbard 1964). No information is available on amounts used.

4.10.5 Discussion of PCB applications with no known commercial use

The possible incorporation of PCBs in various products is virtually endless. Two major factors prevent documenting other uses: time and quantity.

1. Time. The further back, the fewer records have been retained and are available for recreating the history. In the mid-1970s, when Durfee's report was published, Monsanto had made available production and use records. Monsanto's sales records for different applications only go back to 1957 (Durfee et al. 1976).
2. Quantity. Historic low-volume uses often went unrecorded. Small quantities were often sold through intermediate suppliers and the end-uses were never recorded outside the formulator's records. Some "applications" may have been nothing more than a laboratory batch prepared for test and evaluation.

Over the past four decades, a number of PCB uses have been reported that fall in the category of folklore: there is no evidence of their use and no basis for the assertions, although the applications may have been contemplated by lab scientists or salesmen. In an effort not to propagate unsubstantiated rumors, we do not include folklore here.

5 Conclusions

PCBs were used primarily as electrical insulating fluids in capacitors and transformers and also as hydraulic, heat transfer, and lubricating fluids. PCBs were blended with other chemicals as plasticizers and fire retardants and used in a range of products including caulks, adhesives, plastics, coatings, and carbonless copy paper. In the USA, PCBs were manufactured from 1929–1977, although many products remained in service for decades after their manufacture was terminated.

Capacitors (~50%) and transformers (~25%) were the dominant uses of PCBs. Hydraulic and lubrication fluids made up about 6%. The applications where PCBs were incorporated in other products were all minor: NCR Paper was <4% and the numerous plasticizer applications were about 9%.

This article reviews the historic uses of PCBs and discusses, where possible, the relative sales volumes. Especially with smaller volume, military, and third-party uses, documenting a use and/or differentiating between a legitimate commercial use and an experimental test batch is not possible. A major contribution here is to sort out those reported uses which can be documented from those which cannot. The latter category includes probable uses which are not documented.

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