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**Emissions of Polychlorinated Biphenyls as
Products of Incomplete Combustion from Incinerators**

P.M. Lemieux, C.W. Lee, J.D. Kilgroe, J.V. Ryan

U.S. Environmental Protection Agency

Air Pollution Prevention and Control Division

Research Triangle Park, NC 27711

ABSTRACT

Polychlorinated biphenyls (PCBs) have been widely used in the past as industrial chemicals, particularly as additives in electrical transformer cooling oil. Growing evidence of PCBs' role as a persistent, bioaccumulative, human carcinogen has led to the banning of the production and use of PCBs as an industrial chemical in major industrialized countries including the United States. PCBs, however, are still being released into the environment as an unwanted by-product of combustion processes, particularly those associated with chlorinated materials. A subset of PCBs, the coplanar isomers, exhibit biological activity similar to that of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDDs/PCDFs), a widely recognized by-product of combustion processes. Significant progress has been made over the last 10 years investigating the fundamental PCDD/PCDF formation mechanisms, while emissions of PCBs from combustion devices have not been extensively investigated. This paper presents background information on some of the combustion sources that generate PCBs.

INTRODUCTION

A number of persistent organic pollutants (POPs) are of national and international concern due to their persistence, their mobility, their ability to bioaccumulate, and their potential impacts on the health of humans, wildlife, and fish. Much of this concern involves 12 chemical classes that include polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins and -furans (PCDDs/PCDFs), and pesticides such as dichlorodiphenyltrichloroethane (DDT), chlordane, and heptachlor [Wania and MacKay, 1996]. The three chemical classes that are perhaps of greatest concern are PCDDs, PCDFs, and PCBs [U.S. EPA, 1994; U.S. EPA, 1997].

Formation and control of PCDDs/PCDFs from combustion sources have been a public environmental health concern because of reputed carcinogenic, teratogenic, endocrine-disrupting, and persistent and accumulative behavior of these compounds in biological systems. Of the 208 tetra-octa CDD/CDF isomers, 17 (those substituted in the 2,3,7,8 positions) are recognized for their toxicity.

From the 1940's to the 1970's, PCBs were widely used commercial chemicals. They were used primarily as heat transfer fluids and as pesticides. At one time, eight mixtures of PCBs were sold in the U.S. under the trade name Aroclor. Many other mixtures were manufactured elsewhere. In 1977 Congress passed the Toxic Substances Control Act (TSCA), providing EPA with broad "cradle-to-grave" regulatory authority for almost all existing and new chemicals manufactured, imported, or used in the U.S. [Shifrin and Toole, 1998]. In January 1978, EPA banned the manufacture, processing, distribution, or use of any PCBs in any manner other than entirely enclosed. However, the previous manufacture and use of PCBs produced a negative legacy. In 1991, EPA's Superfund Office estimated that PCBs were a major contaminate, accounting for about 34 million cubic yards of material at 20 % of the sites on the National Priorities List [Shifrin and Toole, 1998].

Certain polychlorinated biphenyl (PCB) isomers exhibit similar toxic effects as PCDDs/PCDFs. PCBs substituted with zero or one chlorine atom in the 2,2' or 6,6' (ortho) position on the phenyl ring and one or more meta (m) or para (p) chlorines on each ring (see Figure 1) can assume a planar configuration, leading to a molecule that is similar in structure and orientation to 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD). These coplanar PCBs are also termed “dioxin-like” PCBs.

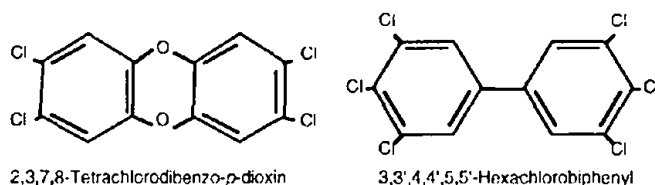


Figure 1. PCBs and Dioxin Structure

Similar to PCDDs/PCDFs, each coplanar PCB isomer has been assigned a toxic equivalency factor (TEF) relative to 2,3,7,8-TCDD [Ahlborg et al., 1994], which has been arbitrarily assigned a value of 1 as shown in Table 1 [NATO, 1988]. By summing the products formed by multiplying the concentrations of the various molecules for which a TEF has been assigned by its respective TEF, a toxic equivalency (TEQ) concentration is derived. The TEQ is used as an estimate of the total dioxin-like toxicity of a complex mixture. Human exposure to dioxin-like PCBs in the environment has been suggested to be very significant in a recent review [Alcock et al., 1998], which showed that the human body burden of dioxin-like PCBs is between 50 and 70 % of the total TEQ, based on published European and North American studies. The review also suggested that dioxin-like PCBs dominate the total TEQ (up to 90% total TEQ) of several major food sources such as fish, oils, and fats consumed by humans.

Efforts to control the risks from PCBs have concentrated on exposure related to the manufacture and use of PCBs, with major emphasis on manufacturing process residues and wastewater discharges. It is generally believed that emission of PCBs from combustion sources results primarily from the incomplete destruction of wastes containing PCBs [U.S. EPA, 1997]. Few people, except combustion experts or individuals in the incineration industry, understand that PCBs can be synthesized during the combustion process via gas-phase or heterogeneous reactions.

Table 1. Toxic Equivalency Factors for PCBs [Ahlborg et al., 1994]

Type	Congener		TEF
	IUPAC No.	Structure	
Non-ortho	77	3,3',4,4'-TCB	0.0005
	126	3,3',4,4',5-PeCB	0.1
	169	3,3',4,4',5,5'-HxCB	0.01
Mono-ortho	105	2,3,3',4,4'-PeCB	0.0001
	114	2,3,4,4',5-PeCB	0.0005 ^{a,b}
	118	2,3',4,4',5-PeCB	0.0001
	123	2',3,4,4',5-PeCB	0.0001
	156	2,3,3',4,4',5-HxCB	0.0005 ^b
	157	2,3,3',4,4',5'-HxCB	0.0005 ^b
	167	2,3',4,4',5,5'-HxCB	0.00001 ^a
	189	2,3,3',4,4',5,5'-HpCB	0.0001 ^a
	Di-ortho	170	2,2',3,3',4,4',5-HpCB
180		2,2',3,4,4',5,5'-HpCB	0.00001 ^a

a – Based on very limited data

b – IUPAC 114, 156, and 157 are expected to have similar TEF values based on similar responses. Although the data are limited, the determination of TEFs for these congeners is supported by their structural similarity.

There are very little available data reporting emissions of PCBs from combustion sources, since PCBs were extensively used as industrial chemicals, and their air emissions as trace combustion by-products were largely ignored [Brown et al., 1995]. Combustion sources in the U.S. do not typically require PCB emissions testing. Only in the Netherlands was a national inventory of dioxin-like PCB sources developed [Alcock et al., 1998]. Based on these limited data, PCBs appear to contribute significantly to the total TEQ emissions from combustion sources. For some sources, such as municipal waste combustors (MWCs), PCBs contributed a relatively small fraction (<5%) of the TEQ [Alcock et al., 1998]. This observation on MWCs was duplicated by some Japanese researchers [Kawakami et al., 1993]. For other sources, however, such as cement kilns, PCBs contributed up to 60% of the TEQ [Alcock et al., 1998]. Metal reclamation and sintering plants have also been identified as an important source of coplanar PCBs [Boers et al., 1994]. PCB TEQs from combustor emissions appear to be dominated by the PCB-126 congener, one of the non-ortho PCBs, which also has the highest TEF (0.1) of all PCBs.

The largest body of PCB combustor emissions data from North America exists from some hazardous waste incinerator (HWI) and cement kiln trial burns performed under the Resource Conservation and Recovery Act (RCRA), from EPA's HWI database [U.S. EPA, 1996], and from some MWCs that were sampled during Environment Canada/U.S. EPA's National Incinerator Testing and Evaluation Program [Finkelstein and Klicius, 1994]. Recently developed RCRA guidance documents supporting current HWI regulations specify that certain sources must measure PCBs during emission tests to support risk assessments as part of the permitting process [U.S. EPA, 1998a; U.S. EPA, 1998b].

The limited available data suggest that PCBs may be formed by the same types of reactions that produce PCDDs/PCDFs [Schoonenboom et al., 1995]. PCDDs/PCDFs are generally believed to be formed via several proposed mechanisms: gas-phase formation [Weber and Hagenmaier, 1999]; heterogeneous formation from organic precursors [Gullett et al., 1994]; and *de novo* synthesis from flyash-bound carbon [Stieglitz, 1998]. Both of these latter mechanisms involve heterogeneous reactions with flyash-bound metals (such as copper) serving as catalytic sites for reactions involving various carbon-containing species in the gas and/or solid phases. These reactions occur downstream of the high temperature combustion zone at temperatures ranging from 250 to 700 °C. The formation reactions from organic precursors are believed to occur relatively quickly while the flue gases pass through the downstream regions of the combustion device, with reaction times on the order of seconds, while the *de novo* synthesis is believed to occur slowly, with reaction times on the order of minutes or hours, while the flyash is held up in the particulate control devices.

If both PCBs and PCDDs/PCDFs are generated through similar mechanisms, it would be expected that emissions of PCBs should correlate with emissions of PCDDs/PCDFs. This correlation would be expected to be very strong if there were a common rate-limiting step involved in the formation of both PCBs and PCDDs/PCDFs. This paper will examine that hypothesis.

APPROACH

PCB and PCDD/PCDF measurement data were gathered from various combustion sources in the literature and from risk assessment, trial burn, and risk burn reports from EPA's Regional Offices as well as from EPA's Hazardous Waste Combustor Database [U.S. EPA, 1996]. All data were placed into units of nanograms per dry standard cubic meter, corrected to 7 % oxygen (O_2), with the exception of the Environment Canada data, which were corrected to 12 % carbon dioxide (CO_2). All PCB data reflect total PCBs, and PCDD/PCDF data reflect the total of all tetra through octa chlorinated congeners of PCDD + PCDF. At this point, no TEQ calculations have been made since sufficient information was not available to make those calculations for all of the data sets.

The data are summarized in Table 2. Sources for the data in Table 2 included test points from two different wet process cement kilns, data from several HWIs burning hazardous waste (HW), from a facility burning chemical demilitarization wastes (nerve gas), from a facility decontaminating soil at a Superfund site, and from several different types of MWCs burning municipal solid waste (MSW) or refuse-derived fuel (RDF). Samples taken at similar operating conditions for a single facility were averaged into a single entry in Table 2. However, if multiple operating conditions were sampled during a parametric test or trial burn, then each discrete operating condition was reported as an entry in Table 2. Also note that some of the HWIs included some quantity of PCB in the initial feed. The presence of PCB in the feed was not corrected for in any way.

Table 2. PCB and PCDD/PCDF Data

Facility Type	Feed	PCBs in Feed	Total PCDD/PCDF (ng/dscm)	Total PCBs (ng/dscm)	Reference
Cement Kiln (Wet)	coal/ hazwaste	N	8.65E-01	9.98E+02	Weston, 1997
Cement Kiln (Wet)	coal/ hazwaste	N	1.17E+00	1.60E+03	Weston, 1997
HWI (Rotary Kiln)	soil	N	2.46E+00	1.12E+02	Midwest Research, 1997a
HWI (Rotary Kiln)	soil	N	3.45E+00	6.96E+01	Midwest Research, 1997b
HWI (Rotary Kiln)	hazwaste	Y	6.78E+02	9.39E+02	US EPA, 1996
HWI (Rotary Kiln)	hazwaste	Y	7.28E+02	3.68E+03	US EPA, 1996
HWI (Rotary Kiln)	hazwaste	Y	2.94E+02	5.70E+02	US EPA, 1996
HWI (Liquid Injection)	hazwaste	Y	5.41E+02	1.47E+04	US EPA, 1996
HWI (Liquid Injection)	hazwaste	Y	5.53E+02	1.46E+04	US EPA, 1996
HWI (Rotary Kiln)	demil	N	1.00E+00	1.70E+01	US EPA, 1996
HWI (Liquid Injection)	hazwaste	N	6.00E+00	5.13E+02	US EPA, 1996
HWI (Rotary Kiln)	hazwaste	N	1.00E+00	2.10E+01	US EPA, 1996
HWI (Rotary Kiln)	hazwaste	Y	1.00E+01	1.09E+03	US EPA, 1996
HWI (Rotary Kiln)	hazwaste	Y	2.60E+01	1.92E+03	US EPA, 1996
HWI (Rotary Kiln)	hazwaste	N	5.70E+01	4.32E+02	US EPA, 1996
HWI (Rotary Kiln)	hazwaste	N	6.40E+01	1.73E+02	US EPA, 1996
HWI (Rotary Kiln)	hazwaste	N	2.60E+01	4.03E+02	US EPA, 1996
Cement Kiln (Wet)	coal/ hazwaste	N	1.22E+01	1.74E+03	Radian, 1998
Cement Kiln (Wet)	coal/ hazwaste	N	2.32E+01	1.35E+03	Radian, 1998
Cement Kiln (Wet)	coal/ hazwaste	N	2.72E+01	1.45E+03	Radian, 1998
HWI (Rotary Kiln)	demil	N	6.90E-03	5.40E+00	US EPA, 1998c
HWI (Moving Grate)	demil	N	1.16E-01	1.29E+01	US EPA, 1998d
MWC (Modular)	MSW	N	2.63E+02	5.80E+01	Environment Canada, 1985
MWC (Modular)	MSW	N	1.57E+02	ND	Environment Canada, 1985
MWC (Modular)	MSW	N	2.21E+02	1.26E+02	Environment Canada, 1985
MWC (Stoker)	RDF	N	7.01E-01	1.19E+01	Environment Canada, 1994
MWC (Stoker)	RDF	N	1.49E+00	1.91E+01	Environment Canada, 1994
MWC (Mass Burn)	MSW	N	1.67E+02	4.30E+03	Environment Canada, 1987
MWC (Mass Burn)	MSW	N	6.33E+01	3.00E+03	Environment Canada, 1987
MWC (Mass Burn)	MSW	N	1.56E+02	4.90E+03	Environment Canada, 1987
MWC (Mass Burn)	MSW	N	5.97E+02	7.00E+03	Environment Canada, 1987
MWC (Mass Burn)	MSW	N	5.25E+02	1.70E+03	Environment Canada, 1987
MWC (Stoker)	MSW	N	4.50E+03	3.60E+02	Kawakami et al., 1993
MWC (Stoker)	MSW	N	1.81E+03	1.30E+02	Kawakami et al., 1993
MWC (Stoker)	MSW	N	6.70E+02	1.90E+02	Kawakami et al., 1993
MWC (Stoker)	MSW	N	4.00E+02	1.10E+02	Kawakami et al., 1993

ND - none detected

RESULTS

PCBs were, for most cases, emitted at higher levels than PCDDs/PCDFs. The data from Table 2 are presented in Figure 2, grouped according to three general facility types: cement kilns, HWIs, and MWCs. There appears to be a very distinct trend with increasing PCB emissions as PCDD/PCDF emissions increase. This trend appears to be independent of the facility type, although the data from the cement kilns are found in the upper area of the cluster of data points. The MWC facility data fit in quite well with the HWI data. Overall, PCB emissions exceeded PCDD/PCDF emissions by approximately a factor of 20, and this trend appeared to hold over five orders of magnitude in PCDD/PCDF emissions.

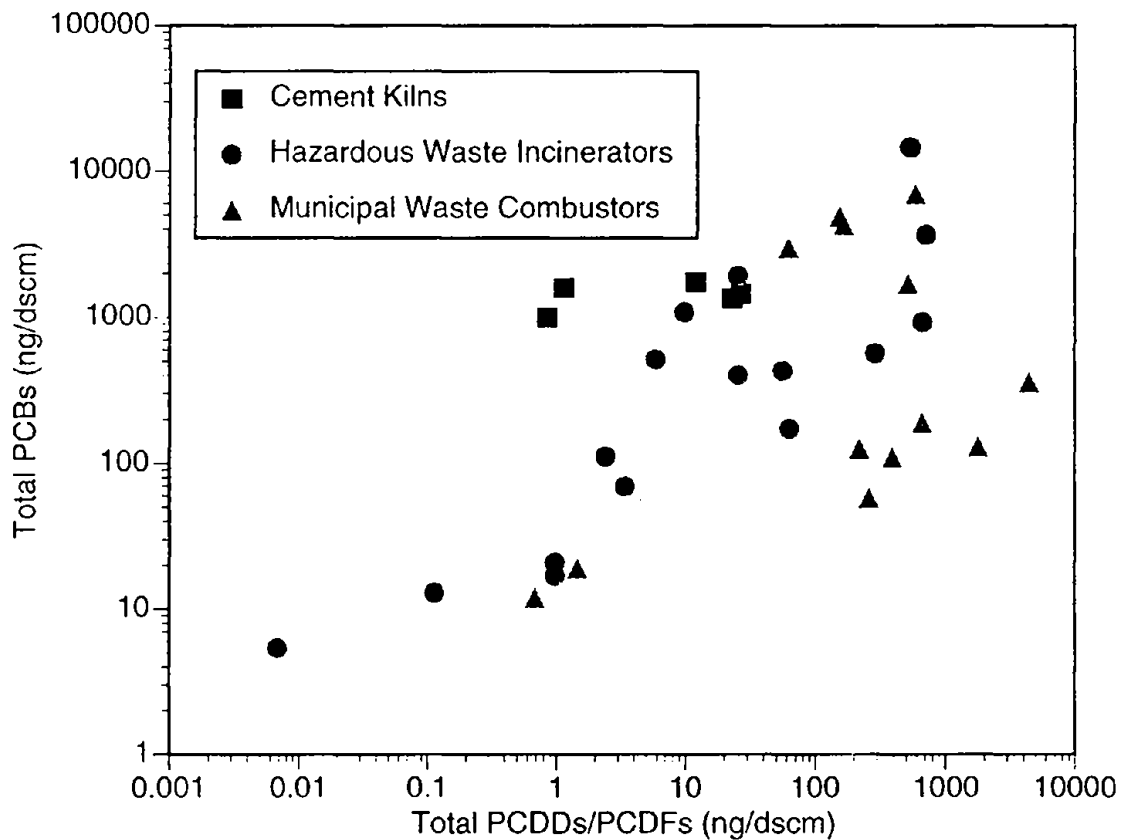


Figure 2. PCDDs/PCDFs vs PCBs

A study in Finland [Kiviranta et al., 1999] compared levels of PCDD/PCDF TEQs and PCBs in human milk and found a similar trend, although the slope of the line was different. In the Finnish study, total PCBs were approximately three orders of magnitude higher than the PCDD/PCDF TEQs. Given that, depending on the source, total PCDDs/PCDFs are nominally a factor of 100 greater than the TEQ, this trend is strikingly similar. The high correlation coefficient ($R^2 = 0.93$) suggested the hypothesis that the sources of PCDDs/PCDFs and PCBs are the same in the Finnish study.

CONCLUSIONS

Data were gathered from various field samples where both PCDDs/PCDFs and PCBs were measured. PCBs were found in the stack in most cases, both in facilities that had PCBs initially in the feed and in those that did not have any PCBs in the feed. Stack concentrations of PCBs were generally higher than stack concentrations of PCDDs/PCDFs, and an apparent trend was observed. This trend was consistent across several orders of magnitude in PCDD/PCDF concentration as well as across several different facility types and feed stocks.

This trend suggests that the same fundamental mechanisms that contribute to the formation of PCDDs/PCDFs may result in formation of PCBs. It also similarly suggests that control strategies intended to reduce emissions of PCDDs/PCDFs may also serve to control PCBs. Although PCBs do not have as high a TEF as 2,3,7,8-TCDD, some of the PCBs have significant TEFs, and since they may be present at concentrations much higher than PCDDs/PCDFs, PCBs may contribute significantly to the overall TEQ for some sources.

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a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Pollution Chlorine Aromatic Compounds Biphenyl Emission Incinerators Combustion Carcinogens	Pollution Control Stationary Sources Polychlorinated Biphenyls Products on Incomplete Combustion Coplanar Isomers	13B 07C 14G 21B 06E
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