

A review of PCB concentrations in tropical media, 1996-2007

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Abstract: A review of publications from the past ten years that report PCB concentrations in sediments and biota is given. During the past ten years only 33 papers out of the published 177 papers on PCBs were from samples obtained in tropical areas. Sediments from the tropics contained relatively low concentrations of total PCBs. The most polluted tropical site in India had sediment concentrations up to 2330 mg/g dw attributed to industrialization and pollution. Sediment from pristine areas were unquantifiable at levels below the detection limits. Biota samples covered a range of species from a range of locations. Biota from pristine tropical areas were unquantifiable at levels below the detection limits. Higher taxa from more developed sites had relatively higher concentrations when compared to other tropical biota, but were still low compared to samples from temperate climates. The lack of data from the tropics still makes correlation of PCB concentrations with other factors problematic. *Rev. Biol. Trop.* 56 (Suppl. 4): 1-9. Epub 2009 June 30.

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Polychlorinated biphenyls (PCB) represent a family of organic chemicals derived from the chlorination of biphenyl and consists of 209 congeners. Commercial PCB formulations were employed in the past mainly as dielectric fluids in power transformers and capacitors, as heat-exchange fluids and in hydraulic machinery, vacuum pumps and compressors. They were also commonly used in paints, lubricants, carbonless copy paper, ink, solvents and insecticides. Their use was banned in many countries in the 1970s; however, existing PCB-containing equipment was used elsewhere for several subsequent decades and may still be in use in some places (Webber 1992).

PCBs are hydrophobic and lipophilic to varying degrees, dependent upon their degree of chlorination. Therefore, their solubility in water is very low, while their solubility in fats and oils is relatively high. Thus, they are found less dissolved in water but rather on the surfaces of solid materials, including sediments and

soils, and also bioaccumulated in organisms (Seba and Snedaker 1995). Since the ocean is the receiving basin for terrigenous freshwater runoff and its entrained materials, some portion of PCBs used in upland areas should eventually reach marine waters. Due to the very properties that made PCBs favorable in electrical applications, these compounds are persistent in the environment. Increasing evidence from laboratory and field studies has shown that trace amounts of many chlorinated hydrocarbons, such as PCBs, in the environment may cause significant endocrine disruption and reproductive disturbance/failure in invertebrates, fish, birds, reptiles and mammals (Wu 1999).

Numerous studies have been conducted since the 1970s on environmental concentrations of PCBs in various media, including air (e.g. Chen *et al.* 1996), water (e.g. Nie *et al.* 2005), sewage sludge (e.g. Cai *et al.* 2007), sediment (e.g. Spongberg 2004a-c), soil (e.g. Zhang *et al.* 2007), and various organisms (e.g.

Spongberg 2006). Fisheries products in several parts of the world (e.g. middle Atlantic Bight of USA) contain unacceptable levels of PCBs (up to 84 $\mu\text{g/g}$ in lobsters and 730 $\mu\text{g/g}$ wet wt. in finfish) (GEASMP 1990). Results of food surveillance programs in Hong Kong from 1995 to 1997 showed that 2.7 ± 7.3 % of shellfish in the market exceeded the local standard (PCB < 2 ppm), posing a significant risk to the marine ecosystems and a health hazard to seafood consumers (Connell *et al.* 1998).

However, not all studies have resulted in such dire conclusions. Most studies are undertaken in areas with a pre-existing environmental concern, such as high population or excessive environmental pollution and degradation, thus skewing the results towards those conditions. By and large the existing literature on PCBs in the environment under-represents pristine or background-type areas and a frequent remark by most authors in their efforts to make global comparisons is that the bulk of work has been done in the middle latitudes where the initial pulse of PCBs entered the environment, leaving the tropics, and to some extent the Southern Hemisphere, less well studied.

Araújo *et al.* (1999) provided an excellent meta-data set of persistent organic pollutant (POP) concentrations in organisms up until 1998. As with other reports, the availability of data from tropical areas was minimal. The peak in total PCB articles occurred in the 1980s, with numbers of articles diminishing since that time. The purpose of this current review is to ascertain the availability and breadth of PCB data from tropical areas published subsequent to the Araújo *et al.* (1999) review and to summarize that data for ease of use by future researchers.

MATERIALS AND METHODS

The data survey was initiated with a computer-generated keyword search of the readily accessible literature and a retrieval of the identified published papers and reports. Only papers sampling environmental media were included, which eliminates all methodological and laboratory studies. All reported data

were accepted without qualification as sample estimates of the target population, and were incorporated in the units used by the authors. This approach ignores the very large variation in analytical procedures used by different analytical laboratories.

Current practice in regulatory evaluation of PCBs in environmental samples involves quantitation as total PCBs or as the total based on Aroclor equivalents (Morrison *et al.* 1996, for example). However, when subjected to processes of degradation and mixing in environmental compartments, the identity of Aroclor mixtures is altered and interpretation of the data is subjective. But, reporting data as total PCBs provides no information about the potential biological significance of the particular mixture of congeners in the sample and may also be misleading since lower chlorinated congeners tend to have lower toxicity. Congeners with less chlorination are more readily metabolized and eliminated and so do not tend to bioaccumulate as much (Hutzinger *et al.* 1974). Data are reported in their original units, such as on dry or wet weight or lipid-adjusted units, to avoid misrepresentation of the original data. A standard method to 'uniformly convert' to common dry-weight estimates can be made by dividing the reported wet-weight values by five i.e., 80 % water, 20 % dry-weight tissue (after Araújo *et al.* 1999). However, since actual water weight is not known, this conversion was not performed for this study. Data presented on a lipid basis is left as reported since the correlation between lipid content and PCB burden is still questionable (Otchere *et al.* 2005, for example).

The most bioaccumulating PCB congeners have five to seven chlorine atoms per molecule. These moderately chlorinated isomer groups consist of 112 different configurations. Since they were synthesized in high proportions in many Aroclor formulations, they tend to be prevalent in environmental matrices. The more highly chlorinated congeners are generally less available to organisms both because they are more tightly bound with soils and sediments and because they usually are present in lower

quantities in the environment. (Goldstein *et al.* 1977, Hutzinger *et al.* 1974). For these reasons MacFarland and Clarke (1989) proposed a compelling argument to report PCB data based on concentrations of individual congeners. As new information about the congeners becomes available, existing data sets can be re-evaluated. MacFarland and Clarke (1989) provide an excellent summary of the environmental occurrence, abundance, and potential toxicity of the different congeners. Therefore, where data are available the dominant congeners are included in this summary.

RESULTS

Characteristics of the data set: The statistics on the articles found published between 1996 and 2007 is listed in Table 1. This list is not complete, but includes all data

easily accessible through normal channels. Dissertations, and articles in more difficult to access journals, are not included. A total of 177 sampling points were identified, including all media types in all climatic zones. This data set is available upon request from the senior author. Using data available from Kottek *et al.* (2006) a climate zone map was overlaid with global positioning coordinate points derived from sampling locations for each study in the searched literature in a Geographic Information System (ArcGIS Desktop, ESRI, Redlands, CA, USA). The Köppen-Geiger climate zone model was used to delineate climatic zone boundaries rather than latitude to represent a more accurate idea of climate zoning at the global level. Average annual precipitation, average monthly precipitation, and average monthly temperature are major inputs to the model. Using data extracted from Figure 1, a

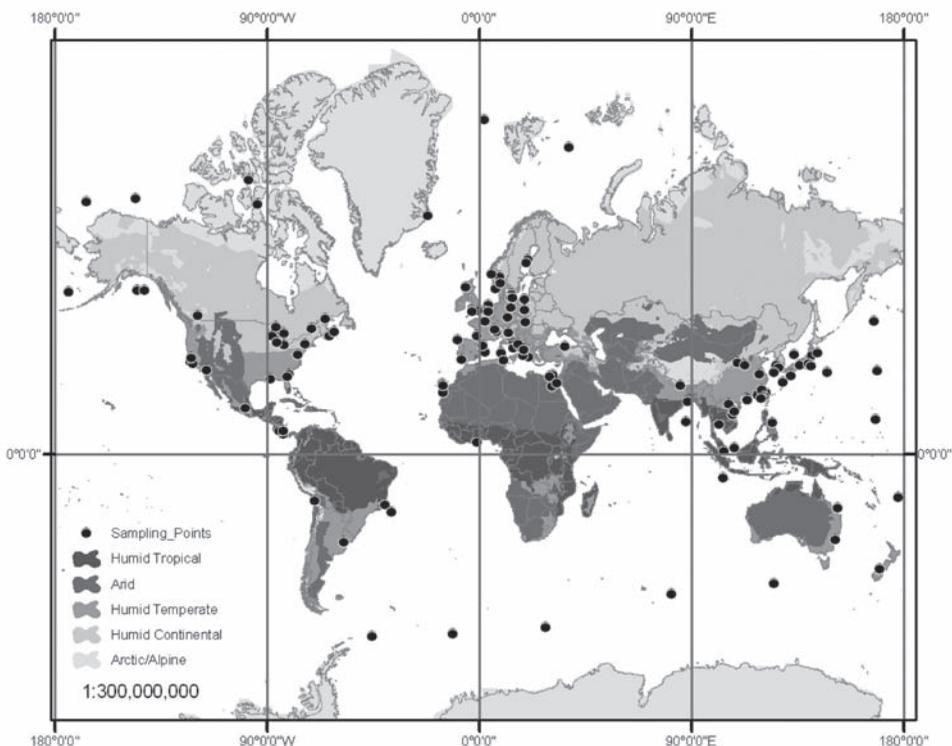


Fig. 1. Map showing distribution of studies around the world from 1996-2007. Shaded color refers to climate zones using the Köppen-Geiger climate classification, as found in Kottek *et al.* (2006).

TABLE 1
Articles found on PCB concentrations in environmental media, 1996-2007

Unique Articles 1996 to 2007	125
Sampling Points 1996 to 2007	147
Adj Sampling Points 1996 to 2007	177**

Year	No. Published	% of total
1996	3	2.4
1997	7	5.6
1998	9	7.2
1999	3	2.4
2000	8	6.4
2001	7	5.6
2002	17	13.6
2003	6	4.8
2004	15	12.0
2005	15	12.0
2006	11	8.8
2007	24	19.2
Total	125	

Climate Zone*	Number of Sampling Points	% of total
Tropical	33	21.0
Temperate	77	49.0
Continental	20	12.7
Arid	8	5.1
Arctic/alpine	10	6.4
Not classifies	9	5.7
Total	157	

* Climate zones use the Köppen-Geiger classification system (Kottek *et al.* 2006)

Type of Matrix	Number of Sampling Points	% of total
Organism	103	58.2
Soil/Sediment	46	26.0
Water	10	5.6
Air	11	6.2
Other	7	4.0
Total	177	

**This number represents number of locations plus amount of matrices analyzed. i.e. 1 sample location, 2 species analyzed equals 2 sampling points.

climate zone was assigned to each sampling point, and subsequent discussion reflects this classification. Thirty-three papers, or 21 % of the total included some samples obtained from the tropics. These studies are represented in Figure 1, with each point representing one study. Although the number of studies conducted in the Tropics has increased in the last decade, the majority of studies (49 %) are still concentrated in temperate zones and in zones of known contamination.

PCBs in soils and sediments from the Tropics: Of the readily accessible papers from the past ten years on PCB research 28 dealt with PCB concentrations in sediment and two with PCB concentrations in soil; eight of these were from tropical regions. Four studies dealt with areas of known pollution in highly industrialized areas. Fu and Wu (2006) studied the effect of precipitation on PCB concentrations in the Er-Jen River in Taiwan. They found that PCB concentrations increased in warmer weather due to higher water temperatures and air/gas exchange rates. Elevated amounts of light fractions after rainfall events did not agree with other studies that attribute this phenomenon to changes in the PCBs due to weathering. Their averaged organic-adjusted sediment PCB concentrations ranged from 769 ng/g dry weight (dw) in May to 1225 ng/g dw in September.

Guzzela *et al.* (2005) studied surface sediments from the highly developed areas of West Bengal, India. Their Total PCB concentrations from ten sites averaged 750 ng/g dw, with the maximum detection of 2330 ng/g dw attributed to proximity to the source. PCBs were dominated by congeners with four to six chlorines.

Frignani (2007) found relatively low total PCB concentrations in the Tam Giang-Cau Hai Lagoon in Central Viet Nam, which correlated to previous work in that country. No non-ortho congeners were detected. Values ranged from 2.03 to 24.5 ng/g dw.

Wurl and Obbard (2005) found a wide variation in total PCBs from sediments around Singapore, ranging from 1.4 to 329.6 ng/g dw (average = 73.9 ng/g dw). High concentrations

were again associated with industry with concentrations decreasing with distance seaward from the sources. PCB concentrations were positively correlated with total organic content in the sediment.

Morrison *et al.* (1996) and Sponberg (2004a-c) provided the only studies that might be considered control studies of more pristine areas. Morrison *et al.* (1996) presents an excellent synopsis of PCB concentrations from the Fiji Islands. The Fiji Islands have little industry and therefore, all sediment samples but one (near a port) showed very low concentrations, mostly less than the detection limits (<10 ng/g dw). The port site had Aroclor 1242/1254/1260 concentrations of 18.11/29.68/20.73 ng/g dw sediment, which is still relatively low.

Sponberg (2004 a-c) conducted an extensive PCB study along the Pacific coast of Costa Rica, including a fairly pristine reef area (Culebra Bay), a deep fjord-type embayment in Golfo Dulce, and the gently sloping estuarine areas of the Gulf of Nicoya. All along the west coast and the port of Limon area along the east Caribbean coast. The average sum of PCB for the all sites was only 2.8 ng/g dw. The Golfo Dulce area had the highest concentrations, averaging 3.6 ng/g dw, even though this area is largely undeveloped. In general, the samples with the highest concentrations were taken from deep, anaerobic waters >100 m, or from the highly developed and polluted port area of Golfito.

In summary, the few studies on PCB concentrations in Tropical sediments show evidence of contamination by nearby activities. However, the values are in general, quite low when compared to studies in temperate climates.

PCBs in biota from tropical regions:

During the past ten years very few studies have been conducted testing the presence of PCBs in the same organisms from different locations. A total of 60 unique articles report PCB concentrations in biota ranging from sipunculan worms and invertebrates, to aquatic mammals such as dugongs, dolphins and whales. Phillips

(1986) noted that organisms can obtain PCBs from several sources, including the atmosphere, water (through either gills or epidermis) and/or food. Differences in PCB content can be due to changes in the surrounding environment that control partitioning between two media, as well as physiological changes in the organism, such as lipid content. Since bivalves tend to have a low level of activity of enzyme systems capable of metabolizing organic pollutants, bivalves have been used as a surrogate to reflect the magnitude of environmental contamination by several researchers.

Seven unique studies were identified from the Tropics, with only two studying similar species (Table 1). Sponberg (2006) studied PCBs in 27 sipunculan worm samples from the west coast of Costa Rica. These worms were chosen because of their ubiquitous presence in coastal waters across the globe. Included in this study was the fairly pristine area of Culebra Bay in the northwest, where the PCB concentrations in the worms were below detection limits. This corresponded with the low levels of PCBs found in the sediments in this area. The Gulf of Nicoya had several worms with PCB concentrations below the detection limits. However, when PCBs were quantifiable, the average sum of PCBs was 22.5 ng/g dw. Golfo Dulce, which had the highest PCB concentrations in the sediments had low to non-detectable PCBs in all samples except one, which had 67.7 ng/g dw. In all cases the congeners were dominated by those with three and four chlorine atoms.

Morrison *et al* (1996) also investigated PCB concentrations in biota from the relatively pristine environment around the Fiji Islands. Riverine mussels, known as kai (*Batissa violacea*) were sampled from 23 sites. All samples were reported as less than the detection limits, although all other organic pollutants were found in levels somewhat higher than in the surrounding sediments.

Cheevaporn (2005) found the sum of PCBs in green mussels (*Perna viridis*) and oysters (*Crassostrea commercialis*) from Thailand to average 1.69 and 4.18 ng/g wet weight. Highest concentration in the sediment was 4.82 ng/g dw.

The higher content in oysters was attributed to a higher lipid content. These values are low for Asia, in general, and are low in comparison to other contaminants found in the vicinity. The bivalves contained mostly PCB homologues with five, six and seven chlorine atoms.

Much higher PCB concentrations were reported by Otchere (2005) in bivalves from Ghana, who found no correlation between the Sum of PCBs and lipid content or body size in cockles (*Anadara senilis*) from Ghana, which was attributed to uptake in food, and not through partitioning from water. A seasonal qualitative difference was noted with concentrations being higher in the dry season with mainly a marine influence compared with the wet, terrestrially-dominated season. Concentrations from Sum of PCBs in cockles from Ghana for the dry and wet seasons averaged 134 and 94 ng/g dw. Oysters (*Crassostrea tulipa*), on the other hand, showed a correlation between sum of PCBs and size, averaging 103 and 117 ng/g dw in the dry and wet seasons, respectively. For many bivalve species the percentage of food ingested with respect to tissue weight increases very rapidly with a decrease in body size. Mussels (*Perna perna*) showed no significant correlations between PCB burden and physiology and averaged 163 and 130 ng/g dw for the dry and wet seasons, respectively. The dominant congeners in all cases were homologues 52 and 28, which have four and three chlorine atoms each, respectively. In general, however, the correlations were confounded by many factors, including the amount and source of food, lipid content, and season.

Strandberg *et al.* (2000) studied biomagnification in amphipods (*Monoporeia affinis*), isopods (*Saduria entomon*) and their predator, the fourhorn sculpins (*Oncocottus quadricornis*) from the Gulf of Bothnia. Lipid-adjusted concentration ranges for amphipods, isopods and sculpin were 670-940, 200-2400, and 890-1100 ng/g, respectively. These values reflected the congener signature of the sediments, with the 6-Cl congeners dominating, followed in order by 5-Cl, 4-Cl, 8-Cl and 3-Cl. Sediments contained an average 9.2 ng/g dw

Sum of PCBs. The low biomagnification in the sculpin was attributed to its low capacity to accumulate PCBs and other POPs and/or the high probability to metabolize or excrete these compounds.

Fu and Wu (2006) noted much higher concentrations in mullet (*Liza macrolepis*) from Taiwan. Lipid-adjusted concentrations averaged 9012 and 8143 ng/g in May and September, reflected the proximity to highly contaminated sites. Although the PCB burden in sediments from this area showed a significant correlation with season, the signature within mullet did not. The larger, more lipid-rich fish had lower PCB concentrations due to the slow rate of PCB incorporation into the lipid as compared to growth rates. The light PCB fraction did increase in the dry season reflecting either gill exchange or bioconcentration from water.

Vetter (2001) analyzed the blubber of dolphins, dugongs, green turtles and a python from northeast Australia. Dolphins (*Tursiops truncatus* and *Delphinus delphis*) contained the highest PCB levels in the study, ranging from 600 to 25,500 ng/g lipid-adjusted. PCBs were found in lower concentrations than were DDTs, females had lower concentrations than males, and overall the data were much lower than similar studies in temperate climates. Congener 153 dominated one third of the sum of the ten major congeners found, followed by 138 and 180, which is typical for other marine mammals. Dugongs (*Dugong dugon*) had lower concentrations, with the highest level found being 209 ng/g lipid-adjusted and many samples with much lower concentrations. The green sea turtle (*Chelonia mydas*) had Sum of PCBs of 171 ng/g lipid. Both the sea turtle and dugongs had PCB levels higher than DDTs. The python (*Morelia spilota*) having a low overall lipid content had many congeners below the quantitation limit and the Sum of PCBs was, therefore, not quantified.

Ueno *et al.* (2003) contributed an excellent collection of organic pollutant concentration data on slipjack tuna (*Katsuwonus pelamis*) worldwide. The livers of all tuna contained PCBs suggesting that these contaminants have

spread all over the world and that the ocean might play a role as a final sink for many POPs. PCBs and DDTs were the predominant compounds with the highest PCB concentrations of 1800 and 1000 ng/g (lipid-adjusted in liver) from offshore Japan and the middle of the North Pacific. DDTs were the predominant class of compounds in tuna samples from the tropical and subtropical regions. Tuna from the southern hemisphere, notably the tropical area off Seychelles, Indonesia and Brazil were significantly lower than other tuna, averaging 120 ng/g (lipid-adjusted in liver), reflecting either their level of usage or environmental assimilation. No significant correlation was found between PCB levels and body weight or body length. Lipid-corrected concentrations were rather uniform within each location. An increase in PCB content with body length in bluefin tuna was attributed to the dietary intake of PCBs and other pollutants in the larger fish being greater than in the smaller skipjack tuna fish. The excretion ratio through gills is slower in larger fish as it has the larger lipid pool for storage (Ueno *et al*, 2002). Skipjack tuna, on the other hand, did not show this trend.

DISCUSSION

A review of the past ten year's studies on PCB concentrations in the environment reveal that concentrations and patterns of organic pollutants in environmental media are influenced by a multitude of interacting factors. PCBs in soil depend on soil taxonomic group, organic matter content and presence of specific vegetation. Sediment concentrations will depend upon the overlying waterbody and drainage basin. Measurements usually also provide only snapshots of the overall situation in space and time. Absolute and relative abundance of PCBs in organisms are subject to even larger confounding factors such as species studied, size and sex, metabolism, age and physiological status. Thus, comparing samples from different latitudes even during the same study can be extremely difficult. The further influence of topography, climate, season and land/ocean

distribution distort clear relationships between latitude, temperature and concentrations.

Wania and Su (2004) used numerical models to aid in the understanding of the "cold condensation" model for the global fractionation process for PCBs and specifically to predict what the compositional changes in space and time are expected to look like if the multitude of confounding factors were to be perfectly controllable. Based on the large variability of air-surface-organic matter exchange properties (vapor pressure, Henry's law constant, octanol-air partitioning coefficient) of the individual PCB congeners within the environmentally relevant temperature range Wania and Su were able to model the global fractionation patterns. They presented a comprehensive study of the global historical fate of several PCB congeners using the Globo-POP model, with a particular focus on changes in the relative composition with latitude and between different compartments.

If PCBs entered the environment at only one point in time and at only one point in space (in the mid-latitudes which is the dominant contributor), they would gradually move from warmer to colder latitudes as they dispersed from the initial entry point into the wider global environment. Due to degradation and other irreversible loss processes that are congener specific, the concentrations gradually are diluted and/or lost from the global environment. The combined effect of these three processes is a peak of chemical moving northward, while at the same time decreasing in peak area and increasing in peak width. Lighter PCB congeners are expected to move to polar regions more quickly, degrade faster and disperse more readily than the heavier ones. But as Wania and Su rightly pointed out, PCBs enter the global environment in many locations, rather than at a single spot over a prolonged period of time.

As intriguing as this model appears, a much larger cache of data from the tropical regions is needed to support this model. Many PCB-containing transformers, etc. have been used in the tropics and little research has been done to document the impact that these

compounds have had on the environment. Data that is readily available by 'normal' search means, to people with limited access to more obscure sources, comprises a meager contribution from tropical research.

The data summarized in this study support the conclusion that PCB levels in the tropics are relatively low when compared to sites in the temperate latitudes. However, the level of PCB usage is so variable as to make conclusions on the fate and migration of PCBs out of or into the tropical environment speculative at best.

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RESUMEN

Se hizo una revisión de los datos publicados sobre concentraciones de PCB en sedimentos y biota durante la última década. Solamente 33 trabajos de un total de 117 sobre PCB fueron de áreas tropicales. Los sedimentos de los trópicos tienen concentraciones relativamente bajas de PCB totales. El sitio más contaminado en la India tenía concentraciones de hasta 2330 mg/g de peso seco y se le atribuye a industrialización y contaminación. En cambio, los sedimentos de áreas no contaminadas mostraron valores no cuantificables, inferiores a los límites de detección. La biota incluye un amplio ámbito de especies de varias localidades. Los organismos de áreas tropicales no contaminadas presentan valores inferiores a los límites de detección. Los táxones de áreas tropicales industrializadas muy contaminadas tienen concentraciones altas cuando se les compara con otra biota tropical, pero aún así las concentraciones son relativamente bajas cuando se les compara con muestras de climas templados. La falta de datos de los trópicos aún dificulta correlacionar las concentraciones de PCB con otros factores.

Palabras clave: PCB, contaminación marina, áreas tropicales, sedimentos.

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