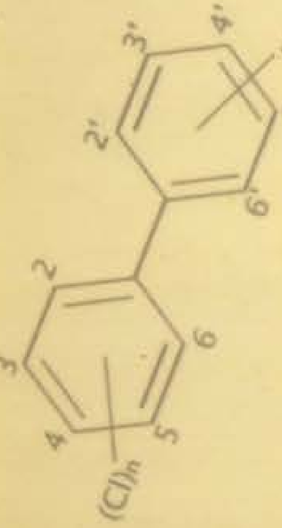
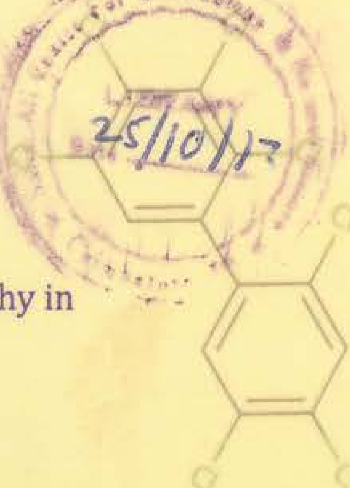
POLYCHLORINATED BIPHENYL (PCB) RESIDUES IN MARINE FISHES AVAILABLE IN COIMBATORE MARKET AND THEIR SUITABILITY FOR HUMAN CONSUMPTION





Ph.D thesis submitted to the

BHARATHIAR UNIVERSITY

Coimbatore

for the award of Degree of Doctor of Philosophy in

ENVIRONMENTAL SCIENCES

by

A. ALAGURAJ

(Reg. No. 2004 R 283)

SACON

Division of Ecotoxicology

Sálim Ali Centre for Ornithology and Natural History (SACON)

(Centre aided by the Ministry of Environment & Forests, Govt. of India)

Anaikatty, Coimbatore – 641 108

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DECLARATION

I, A. ALAGURAJ hereby declare that the thesis, entitled "**POLYCHLORINATED BIPHENYL (PCB) RESIDUES IN MARINE FISHES AVAILABLE IN COIMBATORE MARKET AND THEIR SUITABILITY FOR HUMAN CONSUMPTION**" submitted to the Bharathiar University, in partial fulfilment of the requirements for the award of the Degree of Doctor of Philosophy in **ENVIRONMENTAL SCIENCES** is a record of original research work done by me during **OCTOBER 2004 to OCTOBER 2011** under the supervision and guidance of **Dr. S. MURALIDHARAN, PRINCIPAL SCIENTIST,** Division of **ECOTOXICOLOGY** at **SÁLIM ALI CENTRE FOR ORNITHOLOGY AND NATURAL HISTORY, ANAIKATTY, COIMBATORE - 641 108** and it has not formed the basis for the award of any Degree / Diploma / Associateship / Fellowship or any other similar title of any candidate of any University.

Signature of the Candidate

Signature of the Supervisor

Dr. S. Muralidharan
Principal Scientist, Ecotoxicology
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CERTIFICATE

This is to certify that the thesis, entitled **"POLYCHLORINATED BIPHENYL (PCB) RESIDUES IN MARINE FISHES AVAILABLE IN COIMBATORE MARKET AND THEIR SUITABILITY FOR HUMAN CONSUMPTION"** submitted to the Bharathiar University, in partial fulfilment of the requirements for the award of the Degree of Doctor of Philosophy in **ENVIRONMENTAL SCIENCES** is a record of original research work done by **Mr A. ALAGURAJ** during the period **OCTOBER 2004 to OCTOBER 2011** in the Department of **ECOTOXICOLOGY** at **SÁLIM ALI CENTRE FOR ORNITHOLOGY AND NATURAL HISTORY, ANAIKATTY, COIMBATORE - 641 108** under my supervision and guidance and the thesis has not formed the basis for the award of any Degree / Diploma / Associateship / Fellowship or any other similar title of any candidate of any University.

Countersigned
Head of the Department/ Director

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ACKNOWLEDGEMENTS

I am grateful to my research guide **Dr S Muralidharan**, Principal Scientist, Division of Ecotoxicology, Sálim Ali Centre for Ornithology and Natural History, Coimbatore, for his excellent guidance, support and help till the completion of my work. It was a honour and pleasure to work under him.

I earnestly thank **Dr PA Azeez**, Director, SACON for his support and encouragement. I express my sincere gratitude to **Dr VS Vijayan**, the founder-Director, SACON who provided me necessary facilities to carry out the work. His support and encouragement require special mention.

I also express my deep sense of gratitude to **Drs Lalitha Vijayan, SN Prasad, P Balasubramaniam, S Bhupathy** and **P Promod** at SACON for their help. It is my duty and pleasure to thank **Drs S Chandrasekaran** and **A Avudainayakam** Professor's, TNAU, for their tips in chemical analysis.

I am grateful to **Dr A Gopalakrishnan**, Senior Scientist, NBFGR, Cochin for his help in solving ambiguities in fish identification.

Special thanks are to my friend **Dr V Dhananjayan**, Division of Industrial Hygiene and Toxicology, Regional Occupational Health Centre (South), ICMR, Bangalore, for his advice on statistical tests and interpretation of data which eventually helped me to subsequently write up the thesis.

I thank **Dr R Jayakumar**, **messers P Jayanthi, S Jayakumar, K Ganesan, N Saravana Perumal** and **P Navamani**, Research Scholars, Ecotoxicology, SACON for their help in samples processing and analysis during the study period.

I appreciate **messers S Patturajan**, **R Murugesan** and **T Sivakumar**, Laboratory Assistants for their assistance in the laboratory. I sincerely thank **Mr Subair**, fish merchant (SSR Company), who helped me to collect fish samples from Cochin and Rameshwaram Coast, Tamil Nadu.

I thank **Senior Finance Officer, Finance Officer, Administrative Manager** and other administrative personnel at SACON for their cooperation.

I thank my friends, **MA Rajamamannan, G Srinivas, Vinny R Peter, Menaka, Ananthidevi** and **Suresh Marimuthu** for their help on various occasions.

I thank **Mr V Vaidyanathan,** PA to Director, **Mr Srinivasan**, Computer Assistant, **Mr M Manoharan,** Library Assistant and **Mrs Santhi,** Library attendant for their assistance.

My earnest thanks are to **Mr P Venkatesan** who helped in computer assisted jobs and instrumentation.

Words can never express what I owe to my family members especially my wife **Mrs A Selvarani**, daughter **A Harismita** and son **A Sujay Kiran**, who have been a reservoir of strength, inspiration and cooperation.

(A ALAGURAJ)

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INTRODUCTION

CHAPTER -1 INTRODUCTION

Rapid population growth, enormous urban and coastal development in many of the world's coastal regions have resulted in reduction and depletion of human marine food resources and environmental damages to marine organisms and human health (Aus AID, 1996; Jacinto, 1997; Ciereszko 2001 and Radhakrishnan 2010). The problem of marine pollution due to various types of contaminants such as polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), polyaromatic hydrocarbons (PAHs), petroleum hydrocarbons (PHCs), polychlorinated dibenzo-*p*-dioxins (PCDDs) and heavy metals (mercury, cadmium, arsenic, cobalt and manganese, etc.) has become a global concern. This is further compounded because of the accumulation of the residues of contaminants in the tissues of various species of marine organisms and their biomagnification through the food- chain leading to hazards to human health and environment (Dewailly *et al.*, 1999; Moon and Choi, 2009; Krauthacker *et al.*, 2009).

Fish constitutes an important source of proteins, minerals, vitamins and polyunsaturated fatty acids (PUFAs), especially omega-3 PUFAs. Scientific data indicate that fish consumption reduces the risk of coronary heart disease, decreases mild hypertension and prevents certain cardiac arrhythmias (Kris-Etherton *et al.*, 2002; Perello *et al.*, 2010). At the same time, seafood consumption has been reported as an important route of human exposure to a variety of chemical contaminants (Saeed *et al.*, 1995; Kulkarni *et al.*, 2008; Zhang *et al.*, 2007; Hajeb *et al.*, 2009).

PCBs and brominated flame retardants (BFRs) such as polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs) are industrial chemicals with vast and global use and some of them are still produced in large volumes in India (Devanathan *et al.*, 2009). Due to their high persistent property, these chemicals have been detected in all kinds of environmental and biotic matrices (de Wit 2002). These compounds can accumulate in fat-rich tissues of biota and biomagnify through food web. Particularly, higher trophic organisms including man tend to accumulate high levels of these lipophilic chemicals.

According to Stockholm convention (2001), the member states are suppose to not produce, sell, buy, use, carry through their territories, but to destroy available stocks and to control persistent organic pollutants in the environmental objects. Polychlorinated biphenyls (PCBs) deserved special attention among these pollutants, as they represent a group of highly toxic substances accumulating in the tissues of marine organisms and being conveyed through the food chain to human (Subramanian *et al.,* 2007; Devanathan *et al.,* 2009). PCBs were manufactured from 1929 until their manufacture was banned in 1979 (www.epa.gov/osw/hazard/tsd/pcbs/pubs/about.html 2011) following the passage of the Toxic Substances Control Act (TSCA, 1976). In 1977, when the electrical industry notified Monsanto that suitable replacement fluids were available, Monsanto ceased manufacture of PCBs. Earlier, in 1976, PCBs became the subject of specific regulations under the Toxic Substances Control Act (TSCA), because of their widespread and persistent presence in the environment and concerns about toxicity. In 1978, the United States Environmental Protection Agency (US EPA) issued rules concerning the disposal and labeling of PCBs, and in 1979 it issued rules banning the manufacture of PCBs and placing stringent restrictions on their continued use (US EPA 1987; WHO 1993). But still the amount of PCBs in the global environment has been estimated to be about 37,00,00 metric tons (MT) and further 78,00,00 MT were estimated to be still available in different forms due to various anthropogenic activities (Vit 1992). In India, use of PCBs has been banned in transformers and capacitors since 1997 (Devanathan *et al.,* 2009).

PCBs were never produced in India. Little information is available about the import status of PCB raw materials and availability of finished products in India. However, the estimated quantity of PCBs in India is about 2000 - 4000 MTs (Knight, 1996). It is anticipated that India must have imported PCBs from the countries that manufactures them. Furthermore, the consent for import of PCBs is necessary as from December 12, 1994. One of the sources of release of PCBs is scrapping of merchant ships. It is estimated that scrapping of one ship generates about 0.25 - 0.80 MT of PCBs. (Hess *et al.,* 2001). PCBs containing oils were mostly used in a wide variety of open systems, such as in transformers and capacitors. It is reasonable to assume that this may lead to environmental contamination with PCBs. PCBs have been used in manufacturing of capacitors and transformers. In India, there are number of transformer manufacturers but they do have neither an inventory of the quantity of PCBs being used nor the quantity of PCB-containing equipment. Manufacturers and officials dealing with the transformer oil were not aware about the presence of PCBs in oils. So far Central Power Research Institute, Bangalore (CPRI) has identified 2620 tons of pure PCB and 5000 tons of PCB containing equipment in India. Other sources of PCBs may be used oil from abroad, especially from the Middle East region that includes transformer oil containing PCBs mixed in the general pool (CPRI news, 2010).

PCBs are lipophilic organic compounds whose origin comes from a number of sources. They have a range of toxicity and vary in consistency from thin, light-colored liquids to yellow or black waxy solids. Due to their non-flammability, chemical stability, high boiling point and electrical insulating properties, PCBs were used in hundreds of industrial and commercial applications including electric transformers, capacitors, voltage regulators, switches, reclosers, bushings fluorescent light ballasts, heat transfer and hydraulic equipment as plasticizers in paints, plastics and rubber products including fiberglass, felt, foam and cork, caulking, in pigments, dyes and carbonless copy paper and many other industrial applications (ATSDR, 2008; US EPA 2011).

The PCBs used in these products are halogenated aromatic compounds made up of 209 individual chlorinated biphenyl components, known as congeners differing in the number and position of chlorine atoms along the two phenyl rings and for the physico-chemical properties and toxicological response (Schmidt and Hesselberg, 1992; Storelli *et al.,* 2004; Rawn *et al.,* 2006; USEPA 2011; Mezzetta *et al.,* 2011). They were typically manufactured as mixtures of 60 to 90 different congeners. In the concentrated form, PCBs are either oily liquids or solids with no discernable taste or odor. As the number of chlorines in a PCB mixture increases the flash point rises and the substance becomes less combustible. Also, PCBs with large numbers of chlorines are more stable and thus resistant to biodegradation. The most highly favored PCBs tend to be the ones with large numbers of chlorines. These congeners are also proving to be the ones that present the greatest environmental health risks. (www.environmental chemistry.com/yogi/chemistry/pcb.html, 2011).

Once in the environment, PCBs do not readily break down and therefore may remain for long periods of time cycling between air, water and soil. PCBs can be carried long distances and have been found in snow and sea water in areas far away from where they were released into the environment (USEPA 2011). As a consequence, PCBs are found all over the world. In general, the lighter the form of PCB, the further it can be transported from the source of contamination. PCBs can accumulate in the leaves and above-ground parts of plants and food crops. They are also taken up into the bodies of small organisms and fish (Bocio *et al.*, 2007 and Domingo and Bocio, 2007). As a result, people who ingest fish may be exposed to PCBs that have bioaccumulated in the fish (USEPA 2011).

PCBs are ubiquitous and persistent environmental pollutants with a well known potential toxicity and they were included at the 24th June 1998 United Nations Economic Commission for Europe (UNECE) on Protocol on Persistent Organic Pollutants (POPs) (UNEP, 2008). Although human exposure to PCBs can occur through various routes, food is also the primary source (Domingo and Bocio, 2007). Seafood is a major source of proteins and healthy lipids to humans. Hence, dietary intake of seafood is a major pathway for PCBs (Domingo and Bocio, 2007; Moon and Choi, 2009). Number of studies have shown that the major food sources of PCBs are fat-containing animal products such as fish (Llobet *et al.*, 2003b; Charnley and Doull, 2005; Darnerud *et al.*, 2006; Bocio *et al.*, 2007; Marti-Cid *et al.*, 2007; Bergkvist *et al.*, 2008).

Fishes and other seafoods are bioaccumulators with the highest potential to transfer residues from water to humans (Dorea, 2006a, b). They are also special carriers of much-needed nutrients and functional substances that are crucial to human health. Because of their importance in aquatic ecosystems and in human food sources, fishes are frequently used as standardized testing protocols for purposes such as predicting the bioconcentration factor (BCF) (Barron, 1990), analyzing bioaccumulation in organisms via food chain (Haruhiko *et al.*, 2003; Guo *et al.*, 2008), evaluating human health risk (Dong *et al.*, 2006; Cheung *et al.*, 2007; Dennis, 2007) and monitoring POP pollution (Lanfranchi *et al.*, 2006; Guo *et al.*, 2008).

PCBs may be released into the environment through various transport mechanisms including spills, effluent discharges, incineration or stack emissions and disposal of end-use products (e.g. transformers and capacitors etc.) in dumps and landfills. It has shown that PCBs can cause a wide range of acute and chronic health effects including liver damage, reproductive disorders and skin lesions (NAS, 1979), and they are regarded as suspected carcinogens by the International Agency for Research on Cancer (IARC, 1997). PCB concentrations in the environment are declining but still unacceptably high concentrations are prevalent because of their past widespread application and chemical stability (Schmidt and Heselberg, 1992). In the 1960's, analytical techniques showed PCBs to be ubiquitous and persistent in the environment, with the river and coastal sediments acting as transient reservoirs (Jensen, 1966). The major factors in the dynamics of PCB distribution within surface waters are its low solubility, high specific gravity, and high affinity for solids (Nisbet and Rasmussen, 1992). These result in PCB concentrations in the bottom sediments many times higher than the overlying waters (Dennis, 1975). PCBs can also bioaccumulate to high concentrations in fish tissues even when the concentrations in water are below the usual detection limits (Guo *et al.*, 2008).

PCB contaminants are of particular concern due to their toxicity and ability to bioaccumulate in ecosystem. Several studies have shown that these compounds could exhibit a broad range of toxicological responses including immunotoxicity, reproductive deficits, teratogenicity, endocrine toxicity, carcinogenicity and tumor promotion (Ahlborg *et al.*, 1994 and Ross, 2004). Organochlorine pesticides and PCBs have been detected quite far from the pollution source, because of long-range transport stemming from atmospheric exchange, water currents and animal migration (Zhang *et al.*, 2007). Because of the massive extent of oceanic areas on the earth, water bodies play an important role in the spatial distribution of POPs (Guan *et al.,* 2009). PCBs are susceptible to volatilization from secondary source compartments such as soil, vegetation, water, atmospheric particles and products containing PCBs with temperature as one of the factors controlling the dispersion of PCBs in the environment (Halsall *et al.,* 1995, 1999; Wania *et al.,* 1998). A number of studies on PCBs in the environment are on ambient levels, sources and atmospheric emissions on a regional or global scale (Breivik *et al.,* 2002) followed by studies of PCBs in specific environmental media, such as soil, sediments and water (Guzzella *et al.,* 2005 and Rajendran *et al.,* 2005).

Anthropogenic activities have been accredited for most of the PCBs found in different components of nature, although some studies have indicated that PCBs are being formed due to thermal processes (Ishikawa *et al.,* 2007). Particle-bound PCBs, transported from unknown active sources to urban soil and runoff sediments, however appear to be a major challenge in urban areas (Jartun *et al.,* 2008). Contamination of the environment is apparent not only in areas of direct anthropogenic influence, namely estuaries, owing to the pollutant load in the river run-off but also in remote areas, such as open sea waters, due to long range atmospheric transport and deposition of pollutants. As a result of limited and slow water exchange with the ocean, large freshwater input from rivers and precipitation, and high level of industrialization in catchment areas, the sea is very much under threat from environmental pollution (Potrykus *et al.,* 2003).

Decline in the population of some species of costals birds, such as Common Tern, Forster's Tern, Great Blue Heron and Bald Eagle has been attributed to embryo toxic effects of organochlorine compounds (Elliott *et al.,* 2001). Reproductive deficiencies were noticed at high levels of PCBs in Double-crested Cormorants in the Great Lakes, USA (Cuthbert, *et al.,* 2002). Most of the toxicity was believed to be caused by dioxin-like compounds including coplanar PCBs. The 209 polychlorinated biphenyl (PCB) congeners exhibit a wide range in toxicity to fish, birds and mammals. The accumulated levels of PCBs in migratory birds collected from southern India, one of the wintering grounds for Asian migratory birds have shown to be useful indicators to understand the possibility of PCBs exposure and toxic effects. The migratory birds reflected not only the pollution in the area of sampling but also those in their stopover sites, breeding and wintering grounds (Kunisue *et al.,* 2003).

Until 1980, global production and wide applications of PCBs in different economic sectors was valued at 1.2 to 2.0 million tons. Up to 60 % of previous global production of PCBs penetrated together with industrial and municipal effluents into the seas and oceans, out of which 30 % was accumulated in littoral bottom sediments (Tanabe 1988). They were found in fish, marine mammals as well as in other edible sea resources, such as mollusks, oysters and crabs (Porte and Albaiges 1993; Ciereszko 2001; Fisk *et al.,* 2005; Kannan, *et al.,* 2005; Storelli *et al.,* 2007 and Ramu *et al.,* 2007). Fishes play a main role in human diet; unfortunately they are highly contaminated especially by organochlorine compounds. The most common route of exposure of these contaminants to humans is through consumption of these contaminated fishes (Faroon *et al.,* 2001 and Schantz *et al.,* 2001). Bioaccumulation of these contaminants in fishes has been widely documented all around the world (Cabes *et al.,* 1999; Menone *et al.,* 2000; Vigano *et al.,* 2000; Bocio *et al.,* 2007 and Domingo and Bocio, 2007).

Although there have been controversies surrounding food chain magnification of toxic substances in fishes, many native American tribes were reported to be consuming fish at a rate that exceeds national average (West, 1992). Hence, they were considered to have an elevated risk for the potential hazards of consuming fish containing environmental contaminants such as PCBs, mercury 1,1,1,-trichloro-2.2-bis(p-chlorophenyl)ethane, DDT and their metabolites (ATSDR 2000). A National listing of fish and wildlife advisories indicates that most contaminant studies involve five primary contaminants, namely mercury, PCBs, chlordane, dioxins, and metabolites of dichloro diphenyl trichloroethane (DDT), such as DDE and certain other pesticides bioaccumulating in marine fishes, with continued exposure (US EPA, 2003).

The toxic equivalency factor (TEFs) method serves as an important indirect tool to assess the toxic pollutant load and consequently the risk to exposed organisms. (Ahlborg *et al.,* 1994 and Van den Berg *et al.,* 1998) Speciation of congener specific Polychlorinated Biphenyls (PCBs) in the organisms are essential for a better assessment of the toxic potential. Today there are still unresolved questions about the impact of contaminants on fish-eating bird populations and humans due to chemical residues regularly introduced in the past and still present in the sea and other aquatic ecosystem. The determination of polychlorinated biphenyls in fish species representative of different regions may provide relevant information on pollution sources, transport pathways, spatial distributions and fate in the marine environment.

In India there are studies in the levels of PCBs in human milk, human adipose tissues and human blood (Rao and Banerji 1988; Subramanian *et al.,* 2007 and Devanathan *et al.,* 2009). Moreover Kannan *et al.,* (1992) suggested that the major source of contaminants exist in areas around the aquatic environment and particularly estuarine and coastal regions of India. Therefore, it is of great interest to study the levels of residual quantities of PCBs in fishes consumed by the local population. It has been shown that among food stuffs, fish and fish products have the highest POP concentrations (Kannan *et al.,* 1992; Renterghem *et al.,* 2002; Kannan, *et al.,* 2005; Loutfy *et al.,* 2006; Storelli *et al.,* 2007 and Ramu *et al.,* 2007). Thus the present study was aimed at assessing PCBs in select species of marine fishes sold in Coimbatore market. Today PCBs can still be released into the environment from poorly maintained hazardous waste sites that contain PCBs; illegal or improper dumping of PCB wastes; leaks or releases from electrical transformers containing PCBs; and disposal of PCB - containing consumer products into municipal or other landfills not designed to handle hazardous waste. PCBs may also be released into the environment by the burning of some wastes in municipal and industrial incinerators.

With a long coastline extending along the mainland and the rich areas surrounding the Andaman and Nicobar Islands and Lakshadweep Archipelago and a fairly wide continental shelf and slope, India has rich and varied marine fishery resources. It consists of different species of fishes, crustaceans and molluscs (Latha Unnikrishnan, 2006). The total length of India's coastline is about 8,129 km. The seas that surround the Indian coasts are parts of the Indian Ocean, which has an area of 74,917,000 km^2 (Latha Unnikrishnan, 2006). The Bay of Bengal, large but relatively shallow embayment of the northeastern Indian Ocean occupies an area of 2,172,000 km^2. It is boarded by Sri Lanka and India to the west, Bangladesh to the north, and Myanmar and the northern part of Malay Peninsula to the east.

Some studies on the occurrence of PCBs in different fish species were reported during last few decades (Jensen, 1966; Tanabe 1988; Schmidt and Hesselberg, 1992; Ahlborg *et al.,* 1994; Subramanian *et al.,* 2007 and Devanathan *et al.,* 2009).

However, these studies were focused locally. Nationwide or on a wider geographical area research data are scarce in India. Moreover, studies focused on distributions of PCBs in market seafood in India are very few. Hence, in this context a study was initiated to document the prevalence of PCBs in the marine fishes caught at Cochin and Rameshwaram coasts and sold at Coimbatore market (Ukkadam) for human consumption.

OBJECTIVES

The present study was carried out with the following objectives

- ✓ To assess the magnitude PCB contamination in select species of marine fishes caught from Cochin and Rameshwaram coasts, and sold at Coimbatore market.
- ✓ To document the variation in the level of contamination among select species of fishes.
- ✓ To evaluate the suitability for human consumption based on contamination level.

REVIEW OF LITERATURE

2.1. Chemical properties

Polychlorinated biphenyls (PCBs) are a family of man-made organic chemicals with a common structure (a pair of benzene rings) that vary primarily in their degree of chlorination. Each single form of PCB is called a congener, and is often identified by number (e.g., PCB 153) (ATSDR, 2000). PCBs in pure form are odorless or mildly aromatic solids or oily liquids, often found in mixtures with other organic chemicals (USEPA, 1996 and ATSDR, 2000).

2.2. Toxicity

Studies documenting the ill effects of PCBs on marine fishes are scattered in this country. No systematic work has been done in India to monitor the residue levels of PCBs in various biological components and evaluate their effects. An effort has been made to bring together some of the relevant information available in India. Only pertinent studies in the international contexts have been referred.

The toxicity associated with PCBs and other chlorinated hydrocarbons were recognized very early due to a variety of industrial incidents (Drinker *et al.,* 1937). A conference about the hazards was organized at Harvard School of Public Health in 1937, and a number of publications referring to the toxicity of various chlorinated hydrocarbons were available before 1940. Interest in PCBs first developed in the mid-1960s when researchers in Sweden found trace amounts of the compounds in fish, wildlife and the environment (Jensen, 1966). Two years later, PCBs were confirmed in United States environmental samples. These discoveries soon gave way to concerns about worker safety and public health, eventually prompting regulators to ban PCB production in the United States and other countries and mandate environmental cleanup efforts (Ross, 2004).

The most notorious incidents involving PCBs remain the accidental food poisonings that occurred in Japan in 1968 (Yusho) and in Taiwan in 1979 (Yucheng). Both involved eating rice oil that had been contaminated with heat degraded PCB-containing fluid from leaky food-processing equipment. The two episodes firmly, albeit mistakenly, planted the perception in the public's mind that the PCBs themselves pose serious health risks. Many of those who had consumed the oil became ill, as did some of the children later born to women who had consumed the contaminated oil during their pregnancies (Rogan *et al.*, 1988; Yu *et al.*, 1991). The symptoms associated with the exposure included various physical (somatic) complaints, low birth weights, chloracne (skin disorders) and excess skin pigmentation (hyper pigmentation), especially of newborns (Kuratsune *et al.*, 1972).

2.3. Persistence

PCBs nonetheless are still with us (EPA, 2000). In addition, they often are characterized as ubiquitous environmental contaminants (Kulkarni *et al.*, 2008; ATSDR, 2008 and Mezzetta *et al.*, 2011), garnering extensive media attention and arousing widespread public concern. The environmental transport of PCBs is complex and nearly global in scale. During the last few decades, widespread contamination and toxic effects of PCBs have become a serious environmental problem (Abbassy *et al.*, 2003). The public, legal and scientific concerns about PCBs arose from research indicating that they were likely carcinogens having the potential to adversely impact the environment and therefore undesirable as commercial products. Despite active research spanning over five decades, extensive regulatory actions and an effective ban on their production since the 1970s, PCBs still persist in the environment and remain a focus of attention (UNEP, 1999).

2.4. Production of PCBs

The only North American producer, Monsanto, marketed PCBs under the trade name Aroclor from 1930 to 1977. These were sold under trade names followed by a 4 digit number. The first two digits generally refer to the number of carbon atoms in the biphenyl skeleton (for PCBs this is 12), the second two numbers indicate the percentage of chlorine by mass in the mixture. Thus, Aroclor 1260 has 12 carbon atoms and 60 % chlorine by mass. An exception is Aroclor 1016, which also has 12 carbon atoms, but has only 42 % chlorine by mass. Different Aroclors were used at different times and for different applications. In electrical equipment manufacturing in the USA, Aroclor 1260 and Aroclor 1254 were the main mixtures used before 1950, Aroclor 1242 was the main mixture used in the 1950s and 1960s until it was phased out in 1971 and replaced by Aroclor 1016 (UNEP, 1999).

Use of PCBs continues in closed systems such as capacitors and transformers and the global production are estimated in the order of 1.5 million tons. The United States was the single largest producer with over 600,000 tons produced between 1930 and 1977. The European region follows with nearly 450,000 tons through 1984. It is unlikely that a full inventory of global PCB production will ever be accurately tallied, as there were factories in Poland, East Germany, and Austria that produced unknown qualities of PCBs (Breivik, 2004). Particular attention has long been given to the pollution by such compounds because of their accumulation in living organisms (Volder and Li 1995). In 1977, when the electrical industry notified Monsanto that suitable replacement fluids were available, Monsanto ceased manufacture of PCBs. Earlier, in 1976; PCBs became the subject of specific regulations under the Toxic Substances Control Act (TSCA), because of their widespread and persistent presence in the environment and concerns about toxicity. TSCA authorized the EPA to issue rules to regulate the manufacture and use of PCBs in the U.S. In 1978, the EPA issued rules concerning the disposal and labeling of PCBs, and in 1979 it issued rules banning the manufacture of PCBs and placing stringent restrictions on their continued use. These organic pollutants are found in the global environment, although the use of some of them has been already restricted in many countries (Bordanjandi *et al.*, 2003).

2.5. PCBs in marine environment

The principal route of PCB transport to the marine environment is from waste streams to receiving waters, downstream movement by means of solution and re-adsorption onto particles and by the transport of the sediment itself, until eventually reaching estuaries and coastal waters. The marine environment appears to be the ultimate and major sink for PCBs (CCME 1999). Monitoring of aquatic pollution can be carried out by means of bio-indicator organisms because lipophilic compounds show a high affinity for lipid (Larsson, 1990). In the aquatic environment, PCBs are usually found in much higher concentrations in sediments than in the overlying water.

PCBs have a high affinity for suspended solids, especially those higher in organic carbon. This is supported by their low water solubility and high octanol/water partition coefficients (calculated Log Kow values range from 3.76 for biphenyl to 8.26 for decachlorobiphenyl) (CCME 1999). PCBs, with the exception of some lower-chlorinated compounds, have low volatility and are soluble in organic solvents, particularly hydrocarbons. Temperatures in excess of 1,000°C are required for their complete combustion. Sorption to sediments is the predominate mechanism removing PCBs from the water column. Individual PCBs vary widely in their susceptibility to biodegradation. PCBs with three or fewer chlorine atoms per molecule can be biodegraded by many organisms, whereas those with 5 or more chlorines are resistant to biodegradation and biotransformation. In general, PCB congeners have a low solubility in water, high octanol - water partition coefficients, bioaccumulation potential, and resistance to biodegradation. The physical and chemical properties of PCBs cause their removal from water by sorption to suspended particles and bottom sediments (CCME 1999).

Consequently, PCBs have been identified as an endocrine disrupting group of substances. Accumulation of PCBs in sediments poses a potential hazard to sediment-dwelling organisms. Environment Canada has issued interim marine sediment quality guidelines and these include a guideline of 21.5 ng/g kg^{-1} (dry weight) of total PCBs and above which effects on sediment-dwelling organisms may occur (http://www.ukmarine sac.org.uk/activities/water-quality/wq11.html, 2011).

2.6. Congeners of PCBs

Direct partitioning or adsorption of these contaminants from the aqueous medium plays the major role in the uptake of lipophilic compounds by organisms in the lower trophic levels (Hoke *et al.,* 1994). Some specific PCB congeners that are structurally related to 2, 3, 7, 8, tetrachlorodibenzo-*p*-dioxin (TCDD) have toxicological properties (Safe *et al.,* 1985). These coplanar congeners, namely 77, 126, and 169, according to IUPAC nomenclature, along with their less toxic mono or ortho - substituted analogs are even considered as a greater threat to terrestrial and marine mammals than polychlorinated dibenzodioxins and dibenzofurans (Kannan *et al.,* 1989). The most toxic congeners, namely 77, 126 and 169 were present in very small amounts in most industrial PCB mixtures (Albro *et al.,* 1981).

The most abundant PCBs in the environmental samples analyzed were PCBs nos. 138 and 153 and it is remarkable that the sum of congeners 123+149 also accounted for a high percentage. Congeners with low chlorination grade are more readily metabolized and eliminated than highly chlorinated congeners (Schulte and Acker, 1974; McFarland and Clarke 1989). PCBs are also known to accumulate along marine trophic chains and higher ranking predators usually present greater body burdens with an increased proportion of heavier or higher chlorinated congeners (WHO, 1993).

2.7. PCBs contamination in biotic compounds

WHO (1993) reported that in birds those fed PCBs through their diet, there was evidence of egg-shell thinning, while in sea mammals, there was evidence of reduced capacity. The main effect is on the implantation of the embryo but there can be physical changes to the female reproductive tract. The tragic depletion of fish stock around the world is a direct reflection of environmental disruption. The pesticides and toxic chemical pollutants like PCBs are mainly responsible for this situation (ESCAP, 1988). Data on chlorinated compounds in the edible fishes are also important from the viewpoint of human health (Pastor *et al.,* 1996). Manufacturing levels increased in response to the electrical industries need for a safer cooling and insulating fluid for industrial transformers and capacitors. PCBs were also commonly used as stabilizing additives in the manufacture of flexible PVC coatings for electrical wiring and electronic components to enhance the heat and fire resistance (Kaley *et al.,* 2006).

Diet is a major source of exposure. PCBs can be found in fish, meat and dairy products and tend to bioaccumulate in animal fats. PCBs are classified by the United States Environmental Protection Agency (USEPA) as B2, probable human carcinogens (USEPA; 1996). PCBs ability to cause tumors in rodents was first reported in mice by Nagasaki *et al.* (1972) and Ito *et al.* (1973). These studies were followed by PCBs reports of tumors in rats (Kimbrough *et al.,* 1975; Schaeffer *et al.,* 1984; Norback and Weltman, 1985). In many instances, study limitations made the interpretation of the whole body of data difficult and imprecise. The rat studies nonetheless formed the basis for the EPA's classification of PCBs as probable human carcinogens (EPA, 1988). Besides the EPA, the International Agency for Research on Cancer (IARC, 1987), the National Toxicology Program (NTP, 2006) and the American Conference of Governmental Industrial Hygienists (ACGIH, 1996) have similarly concluded that sufficient evidence exists to classify PCBs as known animal carcinogens. Scientific evidence further indicates that PCBs can cause a variety of health effects in laboratory animals under defined conditions of exposure, including cancer and effects on the immune system, reproductive system, nervous system and endocrine system. Whether any of these effects are relevant to humans at environmentally realistic exposure levels is doubtful given what is known about the evidence in humans and the differences in exposure (US EPA 2011).

In the Belgium dioxin crisis in chicken and pork in 1999, due to the complexity of the PCBs, seven indicator (or marker) congeners were used to identify the composition of PCBs. These seven congeners were PCB - 28 (2,4,4' - trichlorobiphenyl), PCB - 52 (2,2',5,5'tetrachlorobiphenyl), PCB - 101 (2,2',4,5,5' - pentachlorobiphenyl), PCB - 118 (2,3',4,4',5 - pentachlorobiphenyl), PCB - 138 (2,2',3,4,4',5' - hexachlorobiphenyl), PCB - 153 (2,2',4,4',5,5' - hexachlorobiphenyl), and PCB - 180 (2,2',3,4,4',5,5' - heptachlorobiphenyl), which were predominantly present in most PCB - mixtures and in environmental samples (Capel *et al.*, 1985; Frignani *et al.*, 2001).

PCBs decrease mouse oozyte fertility *in vitro* at levels of 0.1μg/ml and higher (Kholkute *et al.*, 1994). Previous studies proved that PCB concentrations in cod livers ranged over 1.2 - 2.6 μg/g wet weight. This means that cod liver poses higher concern to human health since the above figures exceeded the tolerance level of total PCBs (Falandyz *et al.*, 1992).

2.8. Studies in fishes

In a 1986 - 89 EPA national assessment, fish from 97 % of sites contained 10 ppm PCB or less, while one study showed fish from 74 % of sites contained 1 ppm or less (Kuehl *et al.*, 1994). Fish near some heavily contaminated industrial sites, however, contain much higher levels of PCBs in mussels, oysters and other bivalves (Fensterheim, 1993). Bioaccumulations of these persistent contaminants in fishes causing major ill effects in humans have been widely documented all around the world (Cabes *et al.*, 1999; Menone *et al.*, 2000; Vigano *et al.*, 2000). The highest levels of PCBs including the heaviest congeners were found in carnivorous species of fishes (*Micropogonias furnieri, Melanochromis parallelus, Trichiurus lepturns, Centropomus undecimatis* and *Pomatomus altatrix*). This was probably due to bigger size and higher lipid content (WHO, 1993).

Distributions and concentrations of toxic contaminants (i.e. PCBs, OC pesticides) in fish tissue reflect surrounding water and sediment concentrations (Bright *et al.*, 1995; Brown *et al.*, 1998). Thus fish tissue contaminant levels also serve as another indicator for assessing water and sediment quality (US EPA 2000). PCBs and *p,p'*-DDE concentrations in different species of fishes may in part be explained by the rank of the fish in the food chain, its habitat and mode of feeding but it may also reflect differences in the size or age of individuals as well as lipid content of muscle tissues. Also some studies have suggested that the high levels of chlorinated organic contaminants found in fishes are due to biomagnifications through food chain (Bimbos and Mau, 1986).

Yamaguchi *et al.,* (2003) reported the PCB mean concentrations to be 3.32 ng/g, 1.83 ng/g 1.85 ng/g, 36.6 ng/g, and 64.2 ng/g dry weight in marine organisms, namely Roach, Perch, Pike, Dace and Eel respectively. Kannan *et al.* (1992) reported the mean PCB concentrations (wet weight) to be 3.5 ng/g and 330 ng/g in fishes and prawns respectively collected from different locations in India. Studies of Gerstenberger *et al.,* (1997a) has reported the mean PCB concentrations (wet weight) in fishes, namely Walleye (193.3 ng/g), Pink Salmon (17.3 ng/g), Siscowet Trout (370.3 ng/g), Carp (1,403.9 ng/g) and Whitefish (154.0 ng/g) collected from Lake Superior, USA.

The concentrations of PCB 123 +149 reported by Bordajandi *et al.,* (2003) were 1.14 ng/g in Common Trout and 4.77 ng/g in European Eel and concentration of PCB 138 was 1.04 ng/g in Common Trout and 12.3 ng/g in European Eel. Similarly PCB153 recorded in common Trout was 0.94 ng/g and in European Eel it was 5.93 ng/g. Manirakiza *et al.,* (2001) have done studies on seven fish species, namely *Baulengerochromis microlepis, Chrysichthys sianenna, Oreochromis niloticus, Lates stappersii, Limnothrissa miodon, Stolothrissa tanganyikae* and *Lates angustrifrons* in Lake Tanganyika, Burundi, Africa and reported the maximum concentration in *Baulengerochromis microlepis* with PCB 153 (34.7 ± 5.2 ng/g) followed by PCB 149 (23.9 ± 3.6 ng/g). They also recorded PCB 153 (30.1 ± 5.6 ng/g) and PCB 138 (24.5 ± 3.5 ng/g) in *Oreochromis niloticus* and PCB 149 (24.1 ± 3.5 ng/g), PCB (153 21.5 ± 2.9 ng/g) and PCB 138 (26.9 ± 3.5 ng/g) in *Lates stappersii.* PCBs were detected in all fish species analyzed at levels higher than those of pp' DDE, the most abundant OCP residue in Guanabara Bay fish (Sarkar *et al.,* 2002; **Table 2.1**) .

The permitted levels of PCBs in fish samples suggest an increased health hazard to local fish consumers who routinely consume fishes in Oregon USA (Sethajintanin *et al.,* 2004). Studies have shown that at least 18 and perhaps up to 36 of the 209 congeners have potential for toxicity due to their excessive presence in fish/animal tissue and their demonstrated enzyme induction effects (McFarland and Clark 1989, Tanabe 1988).

Table 2.1: Concentration of PCBs in fishes reported worldwide.

S. No	Name of the country	Name of the fish	Name of the compound	Concentration (ppm)	Author & Year
1	Upper Thames, UK	Eel	Total PCBs (Number not mentioned)	64.2 ng/g	Yamaguchi *et al.*, (2003)
2	India	Fishes	Total PCBs (Number not mentioned)	3.5 ng/g	Kannan et al. (1992)
3	USA	Walleye	Total PCBs (Number not mentioned)	(193.3 ng/g)	Gerstenberger et al., (1997)
		Pink Salmon		(17.3 ng/g)	
		Siscowet Trout		(370.3 ng/g)	
		Carp		(1,403.9 ng/g)	
		Whitefish		(154.0 ng/g)	
4	River Turia, Spain	Common Trout	Total PCBs (23)	1.14 ng/g	Bordajandi et al., (2003)
		European Eel		4.77 ng/g	
5	Spain	Fish & Shell fish	Total PCBs (11)	11864.3	Llobet et al., 2003
6	Belgium	Anguilla anguilla	Total PCBs (10)	604.99	Maes et al., 2008
7	Netherlands	Anguilla anguilla	Total PCBs (7)	426.5 ng/g	Van Der Oost et al., 1996
8	Spain	Sardine	Total PCBs (7)	1.19	Bocio et al., 2007
		Tuna		1.17	
		Anchovy		1.24	
		Mackerel		0.89	
		Swordfish		0.42	
		Salmon		0.87	

		Hake		0.34	
		Red mullet		4.15	
		Sole		0.24	
		Cuttlefish		0.02	
9	Mediterranean	Phycis blennoides	Total PCBs (14)	342 ng/g	Storelli et al., 2008
10	Turkey	Acanthobrama marmid	Total PCBs (16)	3.0 ng/g	Erdogrul et al., 2005
		Cyprinus carpio		0.94	
		Chondrostoma regium		0.39	
		Silurus glanis		3.4	
11	USA	Cat fish	Total PCBs (3)	4691 pg/kg	Schecter et al., 1998
12	Egypt	Mullet fish	Total PCBs (12)	0.79 pg TEQ/g	Loutfy et al., 2006
		Bolti fish		0.315 pg TEQ/g	
13	Red Sea, Yemen	Scomberomorus commerson	Total PCBs (Number not mentioned)	0.3 ng/g	Al-Shwafi et al., 2009
		Crenidens crenidens		0.5	
		Rastrelliger kanagurta		0.7	
		Thunnus albacares		0.4	
		Carchaias palasrras		0.1	
		Himantura uarnak		0.9	
		Caranx sem		0.4	
		Scomberoides commersonianus		0.3	
		Chanos chanos		0.5	
		Lutjanus sanguineus		0.8	
		Rachycentron canadus		0.9	
		Euthynnus affinis		0.7	
		Epinephelus areolatus		1.0	

		Panutirus homerun		0.2	
		Sepia pharnais		0.4	
		Sphyraena jello		0.3	
		Penaeus semisulcatus		0.1	
14	Adan Gulf	Scomberomorus commerson	Total PCBs (Number not mentioned)	0.2	Al-Shwafi et al., 2009
		Crenidens crenidens		0.3	
		Rastrelliger kanagurta		0.4	
		Thunnus albacares		0.2	
		Carchaias palasrras		0.1	
		Himantura uarnak		0.7	
		Caranx sem		0.5	
		Scomberoides commersonianus		0.4	
		Chanos chanos		0.3	
		Lutjanus sanguineus		0.6	
		Rachycentron canadus		0.8	
		Euthynnus affinis		0.6	
		Epinephelus areolatus		0.9	
		Panutirus homerun		0.3	
		Sepia pharnais		0.4	
		Sphyraena jello		0.2	
		Penaeus semisulcatus		0.1	
15	Pearl River Delta, South China	Tilapia	Total PCBs (36)	166 ng/g	Nie et al., 2006
		Mandarin fish		156	
		Big head		131	
		Crucian		97.4	

		Grass carp		91.6	
16	South China	Northern snakehead	Total PCBs (44)	7052 ng/g	Wu et al., 2008
		Mud carp		12390	
		Crucian carp		8338	
17	Lagos Lagoon, Nigeria	Red Belly Tilapia - juvenile	Total PCBs (8)	2.94 mg/kg	Adeyemi et al., 2009
		Red Belly Tilapia – adult		0.63	
		Bonga shad- juvenile		0.56	
		Bonga shad – adult		2.53	
		Bagrid cat fish - juvenile		0.70	
		Bagrid cat fish – adult		0.88	
18	Strait of Georgia	Oncorhynchus tshawytscha	Total PCBs (135)	12.03 µg/kg	Cullon et al., 2009
	Johnstone Strait			10.03	
	Lower Fraser River			5.37	
	Puget Sound			1.35	
	Duwamish River			6.38	
	Deschutes River			4.29	
19	Lagos, Nigeria	Pseudotolithus carienensis	Total PCBs (19)	0.4228 mg/kg	Kelechi, 2012
		Pseudotolithus senegalensis		0.438 mg/kg	

2.9. Impact on human

PCBs have also been detected in human breast milk (Vaz, 1995; Rylander *et al.*, 1998; Kunisue *et al.*, 2004a, b and c; Jarrell *et al.*, 2005). PCBs can remain in sediment for many years and PCBs that persist are often called the "weathered" PCBs. Persistent weathered PCBs often contain more highly chlorinated PCBs (with 6-9 chlorine atoms) than PCBs recently released into the environment. Highly chlorinated PCBs may be more toxic than PCBs with lower chlorination. PCBs in sediment can bioaccumulate in fish (Ramu *et al.*, 2007). The effects of these compounds include impaired reproduction, endocrine disruption, immunosuppression and cancer (UNEP, 1996). PCBs lead to potential adverse effects on animals including human. Maternal consumption of PCB contaminated fish in human has been reported to cause reduced birth weight, diminished head and reduced gestational age (Borrell *et al.*, 2004).

Many of the studies reported cord blood levels, maternal blood levels, children's blood levels or maternal milk levels of PCBs, while others have used fish consumption as a substitute for PCB exposure. Lower birth weight has been reported in some of the infants born to mothers who consumed fish that contained PCBs (Fein *et al.*, 1984). As fish eat other fish or bottom - dwelling organisms, they take on the body burden of PCBs present in their prey. Fish are able to metabolize (break down) some PCBs; those that are not metabolized or excreted accumulate in the fish fatty tissues. The result is bioaccumulation of PCBs. Thus humans who consume certain fish may also accumulate PCBs. For these reasons, fish are monitored for PCB concentration in aquatic environment. Various federal regulatory bodies have established fish consumption guidelines, particularly for those fish known to accumulate a variety of chemicals, namely, PCBs and other persistent contaminants (Bocio *et al.*, 2007).

Some investigators have examined the relationship between PCB exposure and potential human health effects using indirect or surrogate measures of exposure such as fish consumption. For example, there have been PCBs reports of neurobehavioral and developmental effects in children (Jacobson and Jacobson, 1996; Korrick, 2001), memory and learning impairment in older adults (Schantz *et al.,* 2001), endocrine - mediated effects including shortened menstrual cycle and other nonspecific reproductive effects (Mendola *et al.,* 1997). However, some of these studies have serious shortcomings with respect to confirmation and quantification of exposure, a critical facet in the assessment of causality between any suspected or supposed toxic compound and subsequent health effects. For obvious ethical reasons, the types of studies conducted in laboratory animals are not conducted in humans. For relevant human data, we must therefore rely on (a) studies of workers occupationally exposed to PCBs, (b) studies involving large - scale or widespread accidental exposures, or (c) low-level environmental exposure studies, such as those involving individuals who consume food (e.g. fish) that contains PCBs.

The studies of PCB-exposed populations collectively suggest that the only consistent adverse health effects attributable to PCBs in humans are skin effects, including chloracne and other effects related to chronic skin and eye irritation (James *et al.,* 1993). These effects occurred only in worker populations with relatively high skin and/or inhalation exposures or in individuals with high accidental exposures. In contrast, there is little documentation that environmental exposures, the type of exposure relevant for the vast majority of people, are associated with such effects.

Serum concentrations of total PCBs (Aroclor 1260) in human beings were frequently found at levels of 5- 8 µg/l. Regardless of the fact that national guideline levels for these toxic substances in blood have not been established, this may still be considered as a matter of concern for human health (ATSDR, 2000). Exposure to environmental contaminants like PCBs can lead to immunosuppression and increased susceptibility to disease in Salmonids and other fishes (Arkoosh *et al.,* 1998). Immunosuppression in humans may result from even minor exposures to the environmental contaminants. Exposure may also contribute to the severity of disease occurrence and lack of ability to resist infection (Luster and Rosenthal 1993; Dean *et al.*, 1994).

It is well documented that the greatest human exposure to PCBs occurs through the consumption of contaminated fish. It is likely that PCB congeners are capable of being released into the general environment and thus, are able to contaminate local food by means of poorly maintained toxic waste sites, contaminated dwellings and through unacceptably poor sanitation systems found in most native communities in the Russian Arctic. As seen from concentrations of contaminants measured in maternal blood serum, indigenous pregnant women living in coastal areas of the Russian Arctic, show levels of exposure to a group of 'long - banned' pollutants, and in particular to HCB, DDT and PCBs, that are among the highest currently reported for all Arctic indigenous peoples (AMAP, 2002).

PCBs in foods are regulated by the U.S. Food and Drug Administration (FDA), which has established tolerances for PCBs in various food items ranging from commercial fish to infant/junior foods to paper and food packaging materials intended for use with human food. According to FDA studies, the amount of PCBs consumed in the diet has decreased steadily (from almost 7 mg/day in 1971 to less than 0.1 mg/day in 1988 (Shank, 1991).

Therefore thorough monitoring of organochlorine residues is crucial for proper assessment of human exposure to these contaminants through food. Several of these have been found to affect the female menstrual function; dietary exposure to PCBs in the form of Aroclor has been found to increase menses duration in Rhesus Monkeys (Truelove *et al.,* 1990; Arnold *et al.,* 1993). In humans, a higher percentage of women who were exposed to PCBs and PCDFs through consumption of accidentally poisoned cooking oil (the Yucheng accident) reported abnormal menstrual bleeding when compared to unexposed controls (Yu *et al.,* 2000). Furthermore, among women who were exposed to TCDD following an explosion in a chemical plant in Seveso, Italy, higher TCDD concentrations were associated with longer menstrual cycles among women who were premenarcheal but not among women who were postmenarcheal at the time of exposure (Eskenazi *et al.,* 2002). On the other hand, menstrual cycle length reductions have been found among women who had consumed more than one meal per month of PCB contaminated fish from the Great Lakes, as well as for women who consumed this fish for seven years or more (Mendola *et al.,* 1997).

Consumption of contaminated sport fish from the great lakes has been associated with exposure to many persistent lipophilic chemicals including PCBs, PCDDs, and pesticides such as DDE, hexachlorobenzene, and mirex (Falk *et al.,* 1999; Fitzgerald *et al.,* 2001). PCBs, PCDDs, DDE and other pesticides are known to cross the placenta and thereby serve as a prenatal source of exposure. These observations have been observed in wildlife and in laboratory animals, raising concern about human health effects (National Research Council, 1999). Exposure from contaminated fish consumption prior to pregnancy will result in a body burden of these persistent contaminants. In addition, contaminated sport fish meals cause a substantial increase in circulating blood levels of many potentially endocrine - disrupting compounds to levels far above the accumulated background body burden associated with chronic exposure (Humphrey, 1987; 1988). If this acute exposure occurs during a critical window of organogenesis, it may increase birth defect risk. Male fetus may be more susceptible than female fetus due to perturbations in the hormonal milien during pregnancy. Fetal androgens are critical for the normal development of the male urogenital organs (Mendola *et al.,* 2005).The main concern over PCBs is their high bioaccumulation capacity. PCBs are soluble in the lipids of biological systems and therefore tend to be bioaccumulated in fatty tissues. Bioconcentration factors of 2,00,000 and greater have been reported for Fathead Minnows *Pimephales promelas* (Duke 1971 and Neely 1977) and Pink Shrimp (Klein and Weisgerber 1976) and up to 1,000,000 in other organisms. Relatively low concentrations of PCBs in the aquatic environment can result in the accumulation of relatively high PCB levels in biota. WHO (1993) quoted experimentally determined BCFs in various aquatic species, ranging from 200 to 70,000 or more. In the open ocean, there is bioaccumulation of PCBs in higher trophic levels with an increased proportion of higher chlorinated biphenyls in high- ranking predators.

Substances of low biological degradability tend to accumulate throughout trophic levels of the food net. For example, concentrations of total PCB increases with the trophic level. Only concentrations of PCBs in sediments are higher than levels in the subsequent trophic levels. It has been demonstrated that chlorinated dibenzo - *p*- dioxin and dibenzo-*p*-furan congeners accumulate with considerable species differentiation. Contribution of the dioxin - like PCB congeners, namely, 77, 105, and 126 to the total TEQ is substantially greater than that of PCDD/PCDF (even in cases of known PCDD/PCDF contamination). In all cases an exchange between trophic levels (sediment < algae < plankton < planktivores < piscivorous fish< piscivorous birds) resulted in an increase of both, total PCB concentration and dioxin - like TEQ (for TEF/TEQ of PCBs) (Fiedler *et al.*, 1994).

Presence of high concentrations of PCBs or their residues in marine mammals has been suggested as the cause for (or contributing towards) pathological changes and reproductive failures in Baltic Seals (Helle *et al.*, 1976), Sealions, Seals and Beluga Whales (Addison 1989); immunity suppression (and hence the possibility of being more susceptible to disease) in harbor porpoises (Kuiken *et al.*, 1994) and Seals (Reijnders 1986; de Swart *et al.*, 1994) changes in the development stability of the Baltic Grey Seal (Zakharov and Yablokov 1990); and premature pupping in California sea lions (DeLong *et al.*, 1973).

It is well documented that many chlorinated organics and other stable compounds are distributed at a global scale through atmospheric transport. The presence of persistent polychlorinated compounds in remote areas such as the Arctic and Antarctic has been reported. PCBs, PCDD/PCDF, HCH, and HCB were found in marine organisms such as Seal Blubber and Pinniped Milk as well as in lake and sea sediments. The occurrence of mainly man - made organochlorines in regions far away from industrialized and densely populated areas indicates that atmospheric transport is an important route to disperse these compounds. All three groups of compounds, namely PCB, PCDD/PCDF, and HCB have the same source areas (densely populated and industrialized regions) (Fiedler *et al.* 1994).

PCBs have been identified in air, snow, ocean water and biota in the marine ecosystem. Although most open uses of these chemicals were curtailed in many industrial countries, a considerable fraction of these compounds is still cycling in the ecosphere. Thus, it was estimated that 20 % of the world production of PCBs, 2,30,000 tons, are present in the upper layers of the ocean, and 790 tons are in the open ocean atmosphere. Large quantities of chlorinated pesticides continue to be used in less developed countries, especially in the Southern hemisphere.

FDA tolerance (action) levels for total PCBs in fish was 2 ppm, the maximum limit established for PCB 153 in fish by the Swedish National Food Administration was 0.1ppm (Atuma *et al.,* 1996). The maximum permissible levels of toxic PCBs recommended by the National Academy of Sciences and National Academy of Engineering (NAS - NAE 1972) (**Table 2.2**), for the protection of aquatic biota are 1000 ng/g to 500 ng/g.

Table 2.2: Human consumption threshold values from different countries (or organizations) around the world.

S. No	Name of the organization	Tolerance levels (ppm)
1	U.S. FDA	2.0
2	Swedish National Food Administration	0.1
3	National Academy of Sciences and National Academy of Engineering	0.5 - 1.0
4	USEPA	2.0
5	Australian FDA	2.0
6	ATSDR	2.0

In India, data are still insufficient to determine the impacts of PCBs on ecological environment. Apart from transformers and capacitors, the ship breaking industry is also a source of PCB release into the environment. A study by Hess *et al.,* (2001) estimates that a typical merchant ship to be dismantled for scrap contains between 250 kg and 800 kg of PCBs, which is found principally in the paint as well as in the vessel machinery. The report further indicated that paints often left on the scrap metal that is re-rolled or re-melted could be another source of PCBs pollution (www.rand.org/publication/MR/MR/1377/MR1377. ch4.pdf 3rd December 2001). Bangalore based Central Power Research Institute (CPRI) has identified 2620 tons of pure PCBs and 5000 tons of PCB containing equipment in India. Other sources of PCBs may be used oil from abroad, especially from the Middle East region that includes transformer oil containing PCBs mixed in the general pool (CPRI news, 2010).

PCBs have been detected in human breast milk from India (Tanabe *et al.,* 1990). PCBs have also been reported in human adipose tissue and blood from India (Rao and Banerji, 1988, 1989). These publications suggest that PCBs are widespread even in developing countries. PCBs and dichloro di phenyl dichloroethane (DDE the most stable metabolite of DDT) are still ubiquitous contaminants in human blood. The source of PCBs to Indians was rather unique, where significant contribution was from cereals and vegetable oils as well as from dairy products. The dietary intake of PCBs, HCB and heptachlor in India are relatively low, which might be due to the minimal contamination of the environment by these compounds (Kannan *et al.,* 1992).

Contamination by PCBs in vegetable oils is greater followed by meat and fishes. When the concentrations were expressed on fat weight basis, fishes were ranked first in PCB concentrations followed by meat and animal fat samples (Kannan *et al.,* 1992). These observations suggest that the major source of PCB contamination exist in areas around the aquatic environment and particularly in estuarine and coastal regions of India. In India, the daily intake of PCBs through the consumption of foodstuffs was estimated at 0.86 μg/person, which is appears to be higher than those of the developed nations (Kannan *et al.,* 1992). Although there is little information on the presence of PCBs in India, these areas are undoubtedly major contributors to the environmental burden with Chlorinated hydrocarbons (Subramanian and Tanabe, 2011). Studies on PCB residues in fishes and their toxic effects on humans are very few. So in order to elucidate accumulation profiles and to evaluate the suitability of fishes as a biomonitor for pollution in the marine ecosystem, and to know the dietary exposure of PCBs to human being, marine fishes in Cochin and Rameshwaram, the study has been organized.

MATERIALS AND METHODS

3.1. Study Area

Coimbatore is an important industrial town in South India. It is located between 10°55' and 11°10' N, and 77°50' and 76°50' E at an approximate altitude of 470 m. Coimbatore is the second largest city by population (0.93 million; Census of India 2011) in Tamil Nadu. The city is a major textile and engineering hub of South India, hence called as Manchester of South India. Coimbatore is situated in the extreme west of Tamil Nadu, near the state of Kerala. It is surrounded by mountains on the west, with reserve forests. The eastern side of the district is predominantly dry. The entire western and northern part of the district border the Western Ghats with the Nilgiri biosphere as well as the Anaimalai and Munnar ranges. The consumption of marine fishes in Coimbatore city is based on the supply from the major coastal regions of South India such as Cochin, Rameshwaram, Kanyakumari, Vijayawada, Mangalapuram and Chavakkad. Of which Cochin and Rameshwaram contribute the maximum (90 %) of marine fishes (**Figure 3.1**) to the local market (Muralidharan *et al.,* 2009). Thus sampling of fishes from the market was restricted to mainly only these two locations.

3.1.1. Cochin

Cochin is located between southwest coast of India at 9°58"N - 76°13"E / 9.967°N - 76.217°E, spanning over an area of 94.88km^2 in Kerala which ranks fourth in the country's total marine fish supply. To the west lies the Arabian Sea and to the east is the urbanized region in the rest of the mainland area. Much of Cochin lies at sea level, with a coastline of 48 km. Cochin features a tropical monsoon climate. Average annual rainfall is 3,228.3 mm, with an annual average of 132 rainy days. Annual temperatures range between 23 and 31°C (73–88°F) with the record maximum being 38°C (100°F), and record minimum 17°C (63°F). The coasts of Kerala are endowed with brackish water lagoons interconnected with natural and man - made canals as also 41 rivers outfalls. This coastal area does receive effluents from industries, domestic, automobiles and oil spills. There are about 300 large and medium scale industries and 1,66,000 small scale industries, most of them are located in the coastal area. Of these 250 large and medium scale industries and 5000 small scale industries do pollute the environment significantly. The effluent load is estimated at 0.15 million metric tonnes per day. Port and defense activities also contribute to the contaminant load (Sharma, 2000). Approximately five million tonnes of oil enter the Arabian Sea each year (ESCAP, 1995). Oil pollution from shipping and offshore oil rigs is also a concern in the East Asian seas.

3.1.2. Rameshwaram

Rameshwaram the coastal town situated at 9°17"N - 79°18"E / 9.28°N - 79.3°E on the east - coast of Tamil Nadu is the gate way to Sri Lanka. It is about 330 km from Coimbatore. It has an average elevation of 10 meters. The religious island spread over an area of 61.8 km^2 happens to be in the shape of a conch. It has dry tropical climate, with average annual rainfall 94 cm, mostly from North East monsoon from October to January. Temperature is around 30^{0}C to 35^{0}C. Highest ever temperature recorded at Pamban station is 37^{0}C and lowest is 17^{0}C. Industries such as sea food processing, leather processing, petro and agrochemicals, cement, mining and automobile are the major sources of pollution along the coastal line of Chennai, Ennore, Cuddalore, Avadayankoil and Tuticorin of Tamil Nadu and Karaikal of Puducherry. Oil transport allows the transports of waste of aquaculture farms are increasingly posing threats to coastal water quality. Approximately, 0.35 million metric tonnes/day of effluent generated through these industrial establishments are let into the sea and the Bay of Bengal receives some 400,000 tonnes from similar sources (ESCAP, 1995).

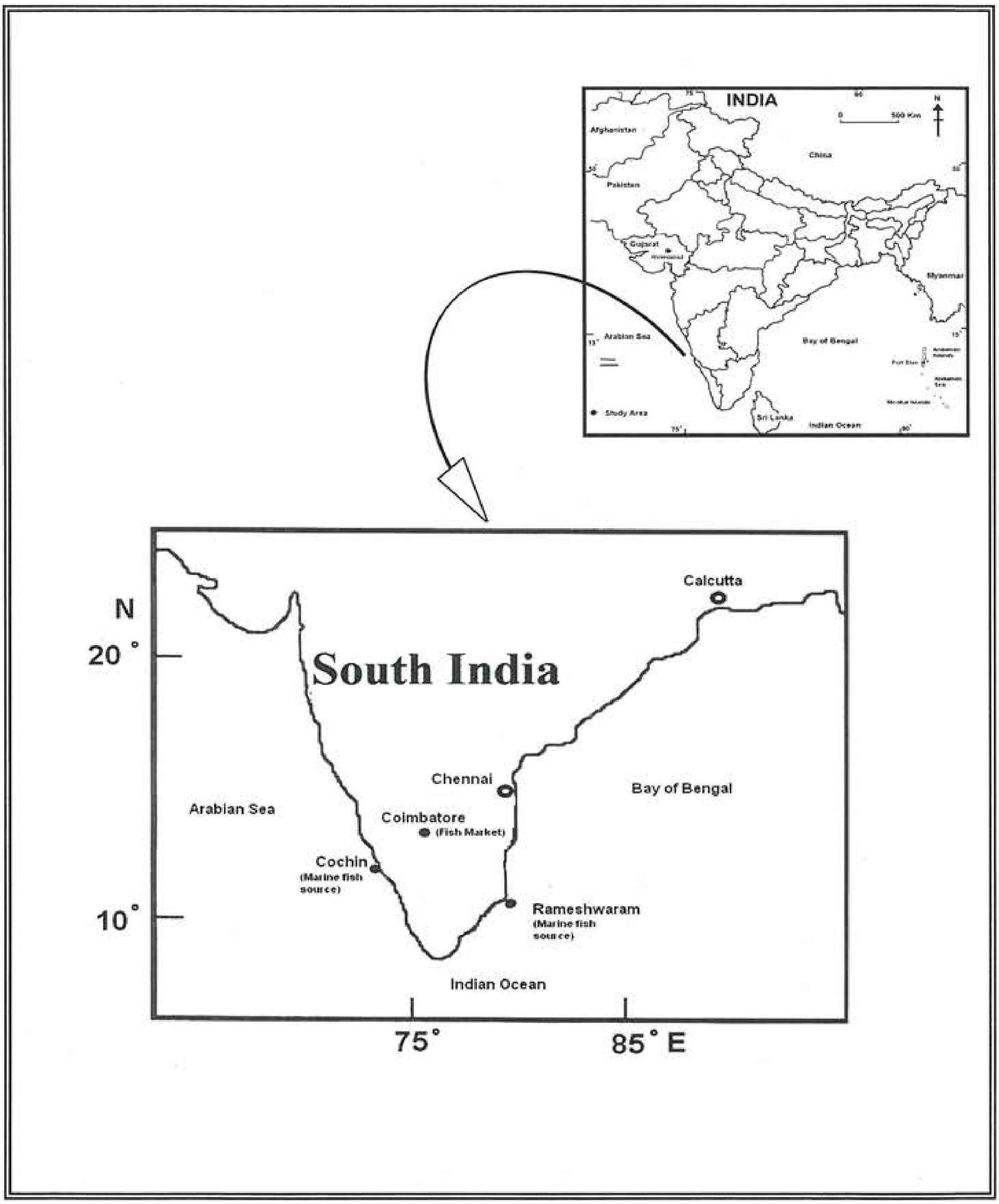


Figure 3.1: Map showing regular supply sites of marine fishes to Coimbatore and study sites

3.2. Sample Collection

A preliminary survey was conducted throughout the Coimbatore city to assess the market potential of marine fishes. Based on the survey, it was found that Ukkadam market acts as a hub for collection/reception of fishes from all the landing centres across the country and its subsequent distribution throughout the city. The survey results showed that about 12 tonnes of marine fishes were sold in the market daily.

The samples from this market were collected from October 2004 to September 2006, i.e., two years which falls into eight quarters with three months being one quarter (October to December 2004 - Quarter 1; January to March 2005 - Quarter - 2; April to June 2005 - Quarter 3; July to September 2005 - Quarter - 4; October to December 2005 - Quarter 5; January to March 2006 - Quarter –6; April to June 2006 – Quarter 7; July to September 2006 – Quarter – 8).

A minimum of three and the maximum of 10 individuals belonging to seven species of fishes, namely Indian Mackerel (*Rastrelliger kanagurta),* Japanese Threadfin Bream (*Nemipterus japonicus*), Oil Sardine (*Sardinella longiceps*), Great Barracuda (*Sphyraena barracuda*), Malabar Travelly (*Carangoides malabaricus*), Tongue Sole (*Cyanoglossus macrolepidotus*) and King Seer (*Scomberomonus commersonii*), which were received at the market from each location namely, Cochin and Rameshwaram was collected during each Quarter. All the seven species of marine fishes were selected on the basis of human consumption and availability throughout the year in both the locations.

Totally 732 fishes were collected during the study period, cleaned of external dirt, labeled and packed in clean polythene bags, transported to the laboratory. The samples were again stored at -20^{0}C deep freezer till the dissection was carried out. Morphometric measurements such as body length and weight were recorded soon after collection and sex of the fish was identified in all possible samples. Common name, length, weight, the major food items and the feeding habits of the fishes are given in the **Table 3.1**.

PLATE 1-FISHES INCLUDED IN THE STUDY

A) Indian Mackerel *(Rastrelliger kanagurta)*

B) Tongue Sole (*Cynoglossus macrolepidotus*)

Plate 1. Continued….

C) Japanese Threadfin (*Nemipterus japonicus*)

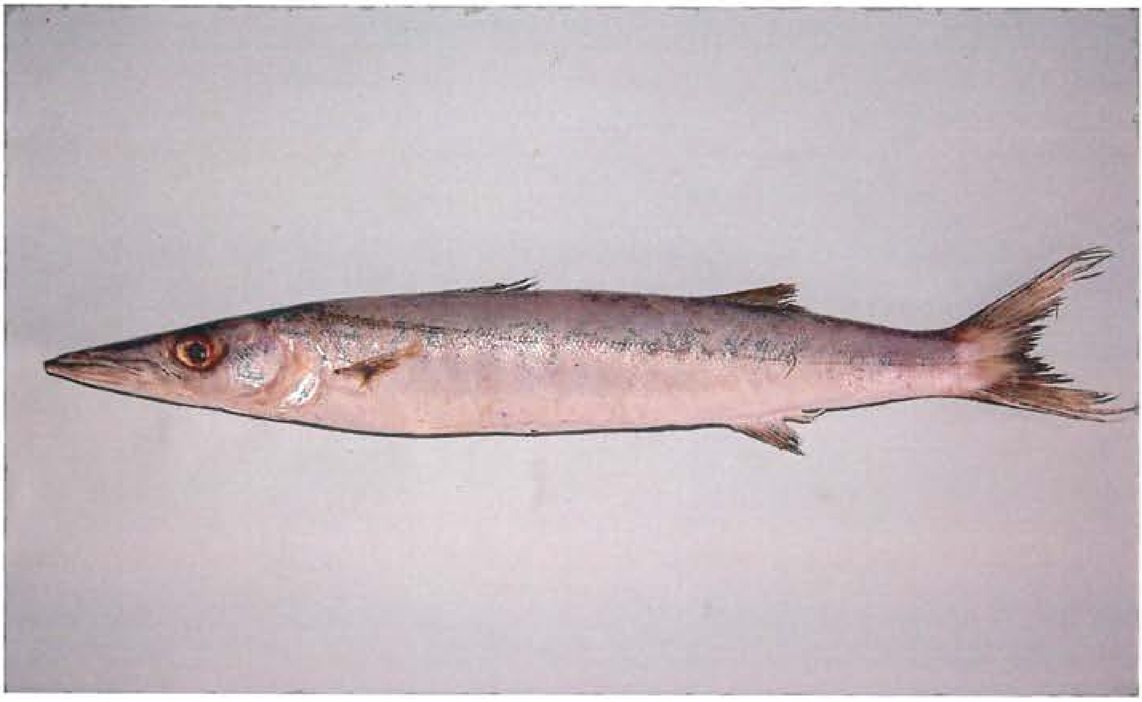
D) Oil Sardine (*Sardinella longiceps*)

Plate 1. Continued....

E) Malabar Travelly (*Carangoides malabaricus)*

F) King Seer (*Scomberomorus commersonii)*

Plate 1. Continued....



G) Great Barracuda (*Sphyraena barracuda)*

H) A view of Coimbatore fish market

Table 3.1: Details of fishes collected from Cochin and Rameshwaram coast between 2004 and 2006.

S. No	Common Name	Scientific Name	Number of Fishes		Length (cm)	Weight (g)	Major food items*
			Cochin	Rameshwaram	Mean ± SD	Mean ± SD	
1	Indian Mackerel	*Rastrelliger kanagurta*	69	61	22.1 ± 1.7	120.1 ± 30.9	Phytoplankton, zooplankton, shrimps and fishes
2	Japanese Threadfin Bream	*Nemipterus japonicus*	77	73	18.4 ± 3.1	73.4 ± 30.7	Small crustaceans, small fishes and invertebrates
3	Oil Sardine	*Sardinella longiceps*	85	80	16.5 ± 1.3	37.3 ± 7.2	Phytoplankton, and small crustaceans
4	Great Barracuda	*Sphyraena barracuda*	30	31	36.1 ± 7.5	247.2 ± 140.5	Fishes, Cephalopods and Shrimps
5	Malabar Travelly	*Carangoides malabaricus*	34	30	22.2 ± 1.9	143.9 ± 30.8	Crustaceans, small squids and fishes
6	Tongue Sole	*Cynoglossus macrolepidotus*	40	47	23.6 ± 5.0	98.8 ± 55.9	Benthic invertebrates
7	King Seer	*Scomberomorus commersonii*	43	32	49.5 ± 11.9	1134.4 ± 872.3	Anchovies, Clupeids, Carangids, Squids, Penaeoid
	Total		**378**	**354**			

A questionnaire survey was conducted from about 325 families across the city to assess the consumption pattern of the marine fishes. The survey aimed at collecting information on preference of the species, family size, frequency of consumption etc., in order to estimate the daily dietary intake of the study species by the local people. The format of the survey form-1 is given below (Muralidharan *et al.*, 2009).

SALIM ALI CENTRE FOR ORNITHOLOGY AND NATURAL HISTORY
Coimbatore – 641 108

SURVEY FORM – 1

SURVEY ON THE CONSUMPTION PATTERN OF MARINE FISHES IN COIMBATORE

Name and address of the respondent :

Occupation :

Income group :

Family size :

Number of members who eat fish in the family :

Frequency of fish consumption :

Amount of fish purchase :

Place of fish purchased :

Most preferred fish :

Indian Mackerel	☐	Japanese Threadfin	☐	Bream Oil Sardine	☐
Great Barracuda	☐	Malabar Travelly	☐	Tongue Sole	☐
King Seer	☐				

Reasons for species specific preference : Taste ☐ Cost ☐ Health ☐

Other information if any

Date

Name of the Surveyor

3.3. Sample Processing

3.3.1. Sample Dissection

Fish samples were taken out from the deep freezer, thawed, cleaned in tap water and scales were sloughed off. Muscle tissue was dissected between the pectoral fin and vent of the fish, minced into smaller pieces and a subsample was taken from the homogenate. About 10 g of the homogenate was weighed using a top loading electronic balance (Mettler AE420) and transferred to clean specimen vials and stored at - 20oC in freezer. Although the vital organs such as gill, kidney and liver are sensitive to organochlorine accumulation, muscle forms the major edible portion in the fish. Therefore muscle tissue alone was analyzed, as the objective is also to determine the dietary intake by the human.

3.3.2 Grinding and Desiccation

About 10 g of the muscle tissue of each sample was taken and mixed with 40 g of anhydrous sodium sulphate. Then it was ground to get a homogenous mixture. The homogenous mixture was then loaded into cylindrical thimble sheet and desiccated over night to remove excess moisture from the sample.

3.3.3. Soxhlet Extraction

The desiccated sample was extracted in soxhlet apparatus for seven hours using 250 ml of hexane and dichloromethane mixture (1:1). The obtained extract was then condensed to a specific aliquot of 5 ml by using the Roto-flask Evaporator (Buchi-R 210). The aliquot was then subjected to acid digestion by adding 2 ml of sulphuric acid. This procedure was carried out to remove the lipid content from the aliquot which may interfere with the chemical analysis.

3.3.4. Column Cleanup

The extracted and acid digested sample aliquot was then purified by column clean up procedure. This involved use of a glass column (25 cm length, 1 cm internal diameter) packed with slurry of silica gel in hexane (60 - 120 mesh). The sample aliquot was placed onto the column and eluted with 100 ml of hexane. The eluant was collected, then condensed to 1 ml with the Roto - flask Evaporator and stored at a -20⁰C deep freezer till the gas chromatography analysis. All the chemicals used in the process were of pesticide residue analysis grade obtained from E - Merck, India.

3.3.5. Chemical Analysis

The identification and quantification of 32 individual congeners **(Table 3.2)** including non-ortho (i.e., PCB 77, 126, 169), mono - ortho (i.e., 105, 114, 118, 156 and189) and di-ortho congeners (i.e., 138, 153 and 180) were performed using Gas Chromatograph (AGILENT Model 6890N - Network GC System) coupled with a Mass Selective Detector (AGILENT Model 5975B - Inert) equipped with a 7683B series auto sampler. PCB 30 (2, 4, 6 Trichloro Biphenyl) was used as Internal standard. MS Spectra for compound identification were produced by both Electron Ionization (EI) full scan and Selected Ion Monitoring (SIM) mode. Column (J&W Scientific Folsom, California, USA) used was a fused silica capillary (30m length x 0.25mm id with 0.25um film thickness of DB - 5 MS (5% Phenyl methyl siloxane). EI full scan (scan range 50 – 600 m/z) was first employed to identify the PCB congeners of interest. Electron ionization was maintained at 70 electron volt throughout the work. Cluster ions were monitored at m/z 222, 256, 290 and 292, 324 and 236, 358 and 360, 392 and 394 to determine the concentrations of di, tri, tetra, penta, hexa and hepta chlorobiphenyls respectively. The GC - MS Tuning (Calibration) was checked before every automated sample set acquition. Tuning was done with FC 43 (Perfluortributlyamine) i.e., heptacosa using m/z 235, 452 and 633 by employing the voltage - adjustment program supplied by the manufacturer for focusing lenses. Column temperature was maintained at 100⁰C for 1 min, subsequently heated to 280⁰C at a rate of 10⁰C /min and maintained for 7 min. The injector temperature was 250⁰C, while the temperatures of the source and transfer line were 250⁰C and 280⁰C respectively. Iolar grade Helium was used as a carrier gas at a constant flow of 1.2 ml/min. Samples were injected in split less mode (split less time, with vent time 1 min) through Digital Pressure and Flow Control (DPFC).

The ionizing energy and EM voltage were 2000 and 70 respectively. Calibration was performed by injecting known amount of standard solutions of Accustandard, USA PCB mix (food and human tissue) by calculating the average relative response factor for each congener. The instrument's performance was continuously monitored by injecting a standard solution before and after a set of six samples. Standard and sample chromatograms are shown in **Figure 3.2 & 3.2a and 3.3 & 3.3a**.

Total PCB concentrations in the sample were quantified by adding the concentrations of individually resolved peaks of different PCB isomers and Congeners. Identification of the PCB congeners relied on the retention time of the peaks, mass fragmentation pattern and NIST Library search factor. Sum PCB concentrations are expressed on a wet - weight (wet/wt) basis except where stated otherwise.

Recoveries of the compounds from fortified samples (100 ng/g) ranged from 94 to 103% and the results were not corrected for per cent recovery and the results were expressed in wet weight basis. Analyses were run in batches of 10 samples plus four Quality Controls (QCs) including one reagent blank, one matrix blank, one QC check sample and one random sample in duplicate. The minimum detection limit for all the compounds analysed was 0.5 ng/g.PCB congeners are referred by their IUPAC numbers throughout this dissertation.

3.3.6. Quality assurance and Quality control

The peaks were qualified and quantified as target compounds and the retention time matched that of the standard compound within 0.1min and the signal-to-noise ratio (S/N) was higher than 3:1. Procedural Blanks were analysed through the whole procedure to check for any interferences and cross contamination arising from chemicals, solvents and glassware. Duplicate samples were analysed for every ten samples. The cod liver standard reference material CRM-349 from BCR, Belgium were analysed for selected PCB congeners.

3.3.7. Statistical Analysis

Statistical analysis of the obtained results was performed with SPSS Software (SPSS for windows, version 10.0). Analysis of variance (ANOVA) was used to test the differences in the concentrations of PCB congeners among species, quarters and between locations. The mean was calculated for the detected values and for values below detectable limit half the value of level of detection was considered. Data were logarithmically transformed to approximate normal distribution before being subjected to analysis of variance (ANOVA) and other analysis. Pearson's rank correlation was applied to evaluate the correlation between chemical concentrations and the length of the fishes. All the statistical analyses were performed at significance level of 0.05 ($p < 0.05$).

Table 3.2: List of PCB congeners included in the study.

S.No	NAME	IUPAC	CAS - NO
1	2,4` - Dichlorobiphenyl	PCB 8	34883 - 43 - 7
2	2,4,4` - Trichlorobiphenyl	PCB 28	7012 - 37 - 5
3	2,2`,5,5` - Tetrachlorobiphenyl	PCB 52	35693 - 99 - 3
4	2,2`,4,5` - Tetrachlorobiphenyl	PCB 49	41464 - 40 - 8
5	2,2`,3,5` - Tetrachlorobiphenyl	PCB 44	41464 - 39 - 5
6	3,4,4` - Trichlorobiphenyl	PCB 37	38444 - 90 - 5
7	2,4,4`,5 - Tetrachlorobiphenyl	PCB 74	32690 - 93 - 0
8	2,3`,4`,5 - Tetrachlorobiphenyl	PCB 70	32598 - 11 - 1
9	2,3`,4,4` - Tetrachlorobiphenyl	PCB 66	32598 - 10 - 0
10	2,3,4,4` - Tetrachlorobiphenyl	PCB 60	33025 - 41 - 1
11	2,2`,4,5,5` - Pentachlorobiphenyl	PCB 101	37680 - 73 - 2
12	2,2`,4,4`,5 - Pentachlorobiphenyl	PCB 99	38380 - 01 - 7
13	2,2`,3,4,5` - Pentachlorobiphenyl	PCB 87	38380 - 02 - 8
14	3,3`,4,4` - Tetrachlorobiphenyl	PCB 77	32598 - 13 - 3
15	2,2`,3,3`,4 - Pentachlorobiphenyl	PCB 82	52663 - 62 - 4
16	2,3`,4,4`,5 - Pentachlorobiphenyl	PCB 118	31508 – 00-6
17	2,3,4,4`,5 - Pentachlorobiphenyl	PCB 114	74472 - 37 - 0
18	2,2`,4,4`,5,5` - Hexachlorobiphenyl	PCB 153	35065 - 27 - 1
19	2,3,3`,4,4` - Pentachlorobiphenyl	PCB 105	32598 - 14 - 4
20	2,2`,3,3`,5,6,6` - Heptachlorobiphenyl	PCB 179	52663 - 64 - 6
21	2,2`,3,4,4`,5` - Hexachlorobiphenyl	PCB 138	35065 - 28 - 2
22	2,3,3`,4,4`,6 - Hexachlorobiphenyl	PCB 158	74472 - 42 - 7
23	3,3`,4,4`,5 - Pentachlorobiphenyl	PCB 126	57465 - 28 - 8
24	2,3,4,4`,5,6 - Hexachlorobiphenyl	PCB 166	41411 - 63 - 6
25	2,2'3,4'5,5'6 - Heptachlorobiphenyl	PCB 187	52663 - 68 - 0
26	2,2`,3,4,4`,5`,6 - Heptachlorobiphenyl	PCB 183	52663 - 69 - 1
27	2,2`3,3`4,4` - Hexachlorobiphenyl	PCB 128	38380 - 07 - 3
28	2,3,3`4,4`,5 - Hexachlorobiphenyl	PCB 156	38380 - 08 - 4
29	2,2`,3,4,4`,5,5` - Heptachlorobiphenyl	PCB 180	35065 - 29 - 3
30	3,3`4,4`5,5` - Hexachlorobiphenyl	PCB 169	32774 - 16 - 6
31	2,2`,3,3`,4,4`,5 - Heptachlorobiphenyl	PCB 170	35065 - 30 - 6
32	2,3,3`,4,4`,5,5` - Heptachlorobiphenyl	PCB 189	39635 - 31 - 9

Note: IUPAC = International Union of Pure and Applied Chemistry
CAS-No = Chemical Abstracts Service

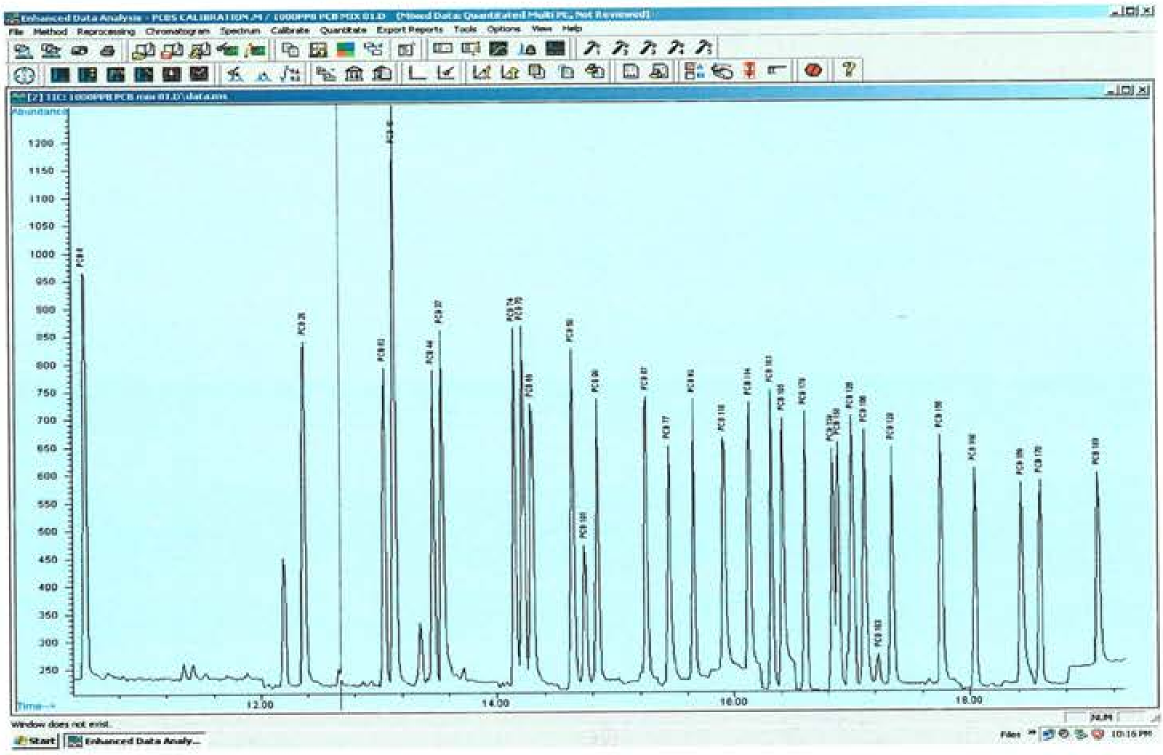


Figure 3.2: Standard chromatogram of 32 individual PCB congeners qualified and quantified in GC-MSD.

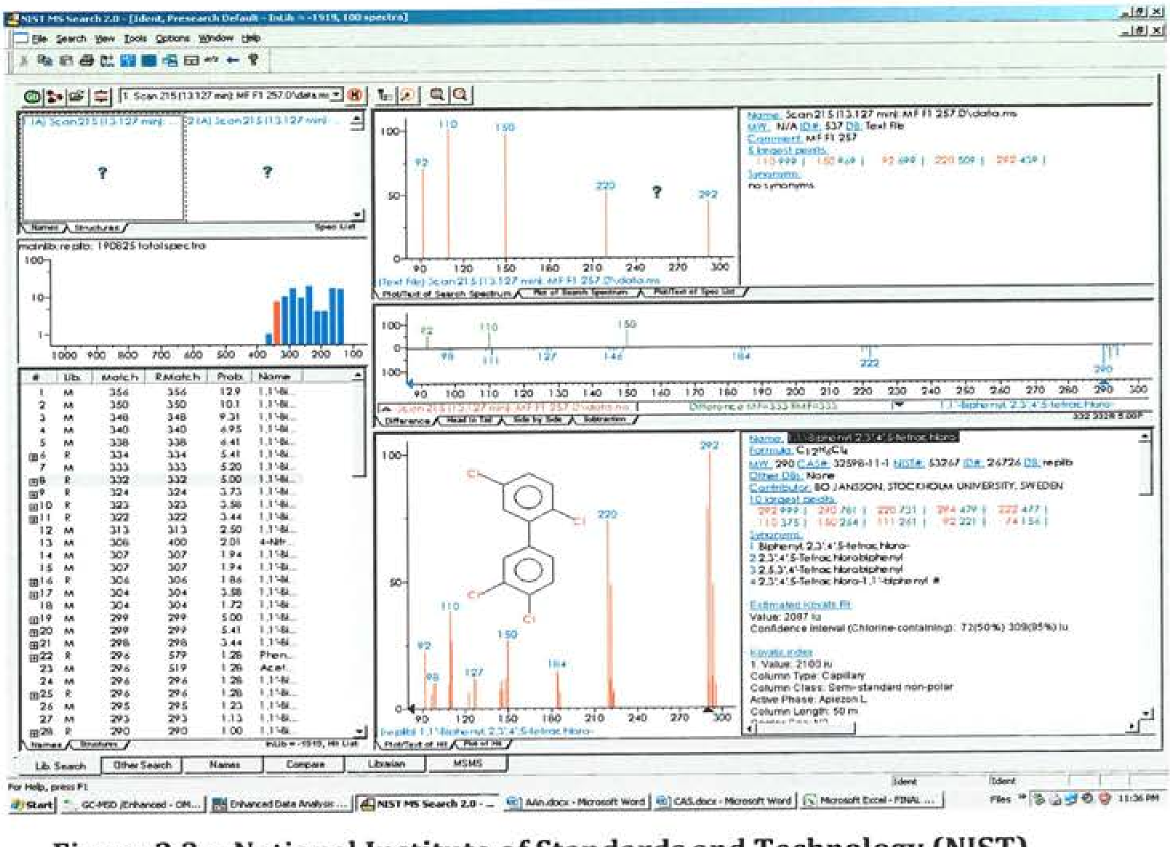


Figure 3.2a: National Institute of Standards and Technology (NIST) library details of PCB congeners standard chromatogram.

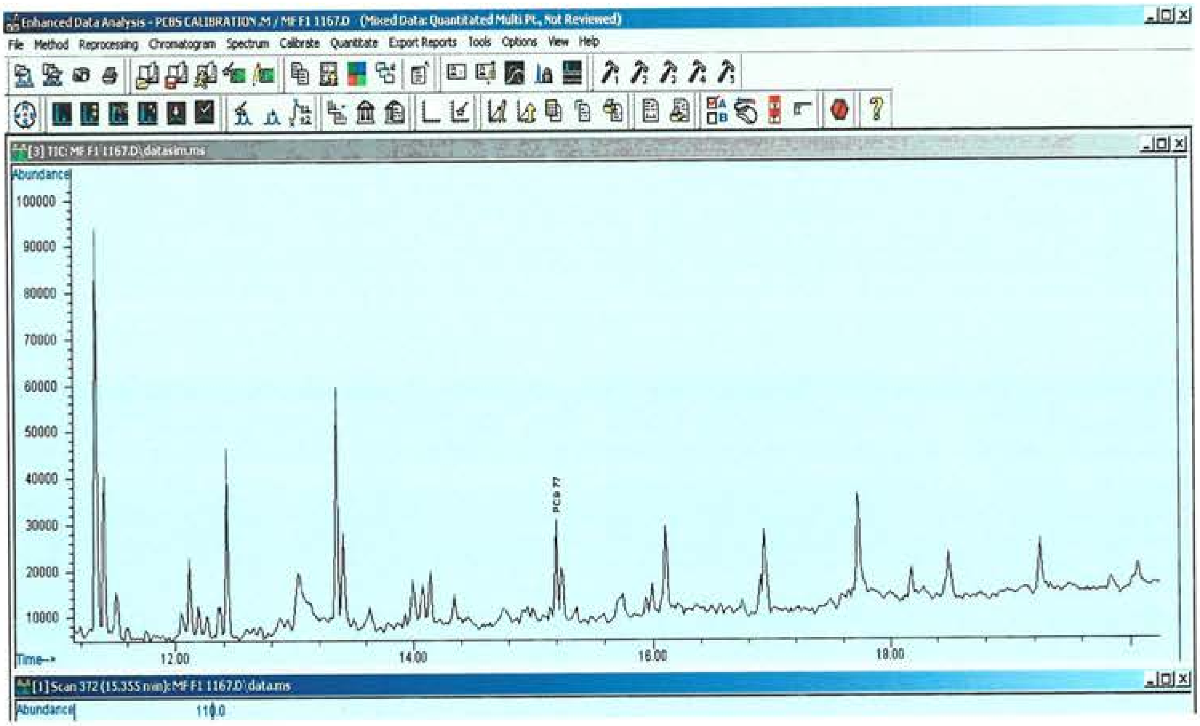


Figure 3.3: Sample chromatogram of 32 individual PCB congeners qualified and quantified in GC-MSD.

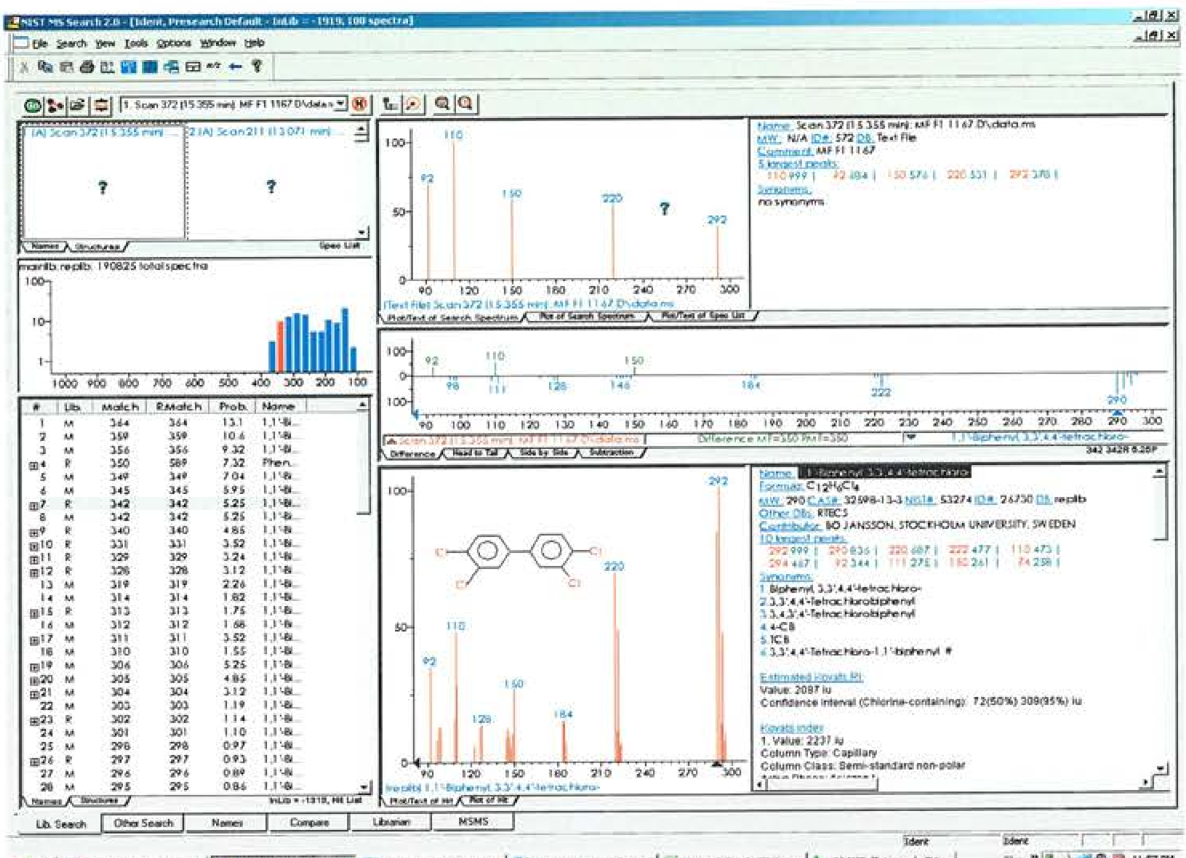


Figure 3.3a: NIST library details of PCB congeners sample chromatogram

RESULTS

4.1. Variations in concentration of PCBs among species of fishes and between locations studied.

Totally 732 samples of muscle tissues comprising seven species of marine fishes from Cochin and Rameshwaram coast collected between 2004 and 2006 were analysed for the residues of 32 individual PCB congeners. The list of species and their weight, length and major food items are presented in **Table 3.1**. Average weight of the fishes included in the present study ranged between 37.3 ± 7.20 and 1134.4 ± 872.3 g. The average length of the fishes ranged between 16.5 ± 1.33 and 49.5 ± 11.9 cm.

Among the seven species of fishes studied, Indian Mackerel *(Rastrelliger kanagurta)* and Oil Sardine (*Sardinella longiceps*) feed on phytoplankton, zooplankton and small crustaceans. Species, namely Japanese Threadfin Bream (*Nemipterus japanicus)*, Great Barracuda *(Sphyraena barracuda)* and Malabar Travelly *(Carangoides malabaricus)* feed on small fishes, shrimps, squids and invertebrates. Whereas, Tongue Sole (*Cyanoglossous macrolepidotus*) and King Seer (*Scomberomonus commersonii*) feed on benthic invertebrates and anchovies, clupeids and carangids respectively **(Table 3.1)**.

Among the fishes collected from the Rameshwaram coast, the maximum concentration of total PCBs was recorded in *Sphyraena barracuda* (29.64 ± 2.14 ng/g) followed by *Carangoides malabaricus* (27.78 ± 1.99 ng/g) and *Rastrelliger kanagurta* (27.06 ± 1.07 ng/g). The minimum concentration of total PCBs was recorded in *Nemipterus japonicus* (22.65 ± 1.99 ng/g) (**Figure 4.1**). Whereas in Cochin, the maximum concentration of total PCBs was recorded in *Rastrelliger kanagurta* (30.88 ± 0.97 ng/g) and *Sphyraena barracuda* (30.68 ± 1.76 ng/g), followed by *Carangoides malabaricus* (26.34 ± 1.67 ng/g) and the minimum concentration of total PCBs was recorded in *Scomberomonus commersonii* (22.87 ± 1.49 ng/g) (**Figure 4.2**).

When fishes of both the locations (Cochin and Rameshwaram) were considered together, concentration of total PCBs was the maximum in *Sphyraena barracuda* (30.15 ± 1.38 ng/g) *Rastrelliger kanagurta* (29.09 ± 0.74 ng/g) recorded the second highest concentration followed by *Carangoides malabaricus* (26.99 ± 1.28 ng/g) and minimum concentration was recorded in *Scomberomonus commersonii* (23.91 ± 1.14 ng/g) (**Table 4.1**). Based on ANOVA, differences in PCB concentration among species were highly significant ($P<0.05$) whereas there was no significant difference observed between locations ($P>0.05$). Hence the samples received from both the locations were treated together for further data analysis.

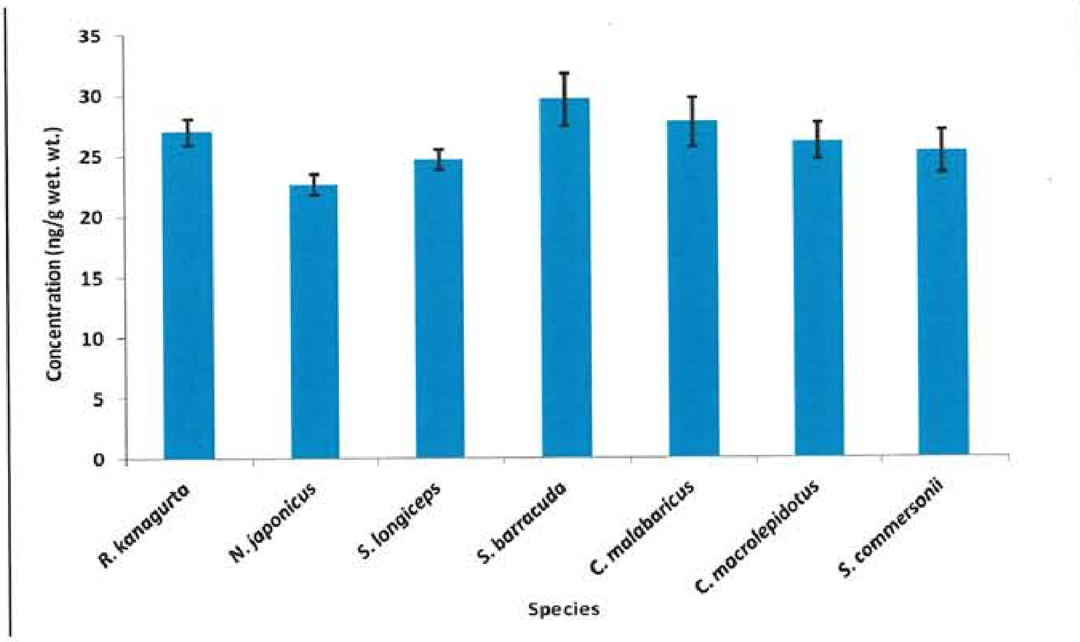


Figure 4.1: Variation in total PCBs levels (Mean ± SE) in the fishes of Rameshwaram.

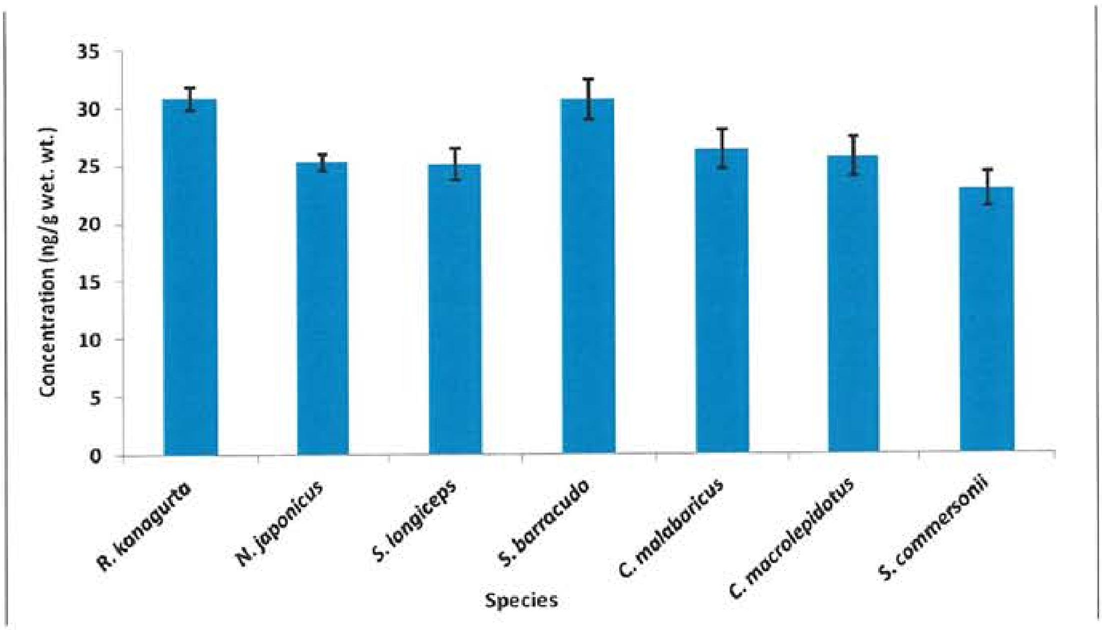


Figure 4. 2: Variation in total PCBs levels (Mean ± SE) in the fishes of Cochin.

Table 4. 1. Total PCBs concentration (ng/g wet wt.) recorded in fishes collected from Cochin and Rameshwaram coast between 2004 and 2006.

Common Name	Scientific name	N	Mean	SE
Indian Mackerel	*Rastrelliger kanagurta*	130	29.09	0.74
Japanese Threadfin Bream	*Nemipterus japonicus*	150	24.13	0.56
Oil Sardine	*Sardinella longiceps*	165	24.88	0.77
Great Barracuda	*Sphyraena barracuda*	61	30.15	1.38
Malabar Travelly	*Carangoides malabaricus*	64	26.99	1.28
Tongue Sole	*Cyanoglossus macrolepidotus*	87	25.99	1.12
King Seer	*Scomberomonus commersonii*	75	23.91	1.14

4.2. Variation in concentration of total PCBs between sex

Variation in total PCB concentrations between sexes was not significant (t - test, $P>0.05$) (**Figure 4.3**). Further, pattern of total PCB load was not similar in all the species analysed. Comparatively higher load of total PCBs was recorded in both male and female *Sphyraena barracuda;* 32.0 ± 1.89 ng/g and 31.32 ± 4.03 ng/g respectively, followed by *Carangoides malabaricus* (28.19 ± 1.66 ng/g in male and 27.18 ± 4.95 ng/g in female), *Nemipterus japonicas* (25.69 ± 0.89 ng/g in male and 22.18 ± 1.23 ng/g in female) and *Sardinella longiceps* (26.21 ± 1.28 ng/g in male and 25.46 ± 1.20 ng/g in female). In contrary to all the species, only *Scomberomonus commersonii* had the maximum PCB burden in female (28.67 ± 1.67 ng/g) than male (26.77 ± 4.06 ng/g).

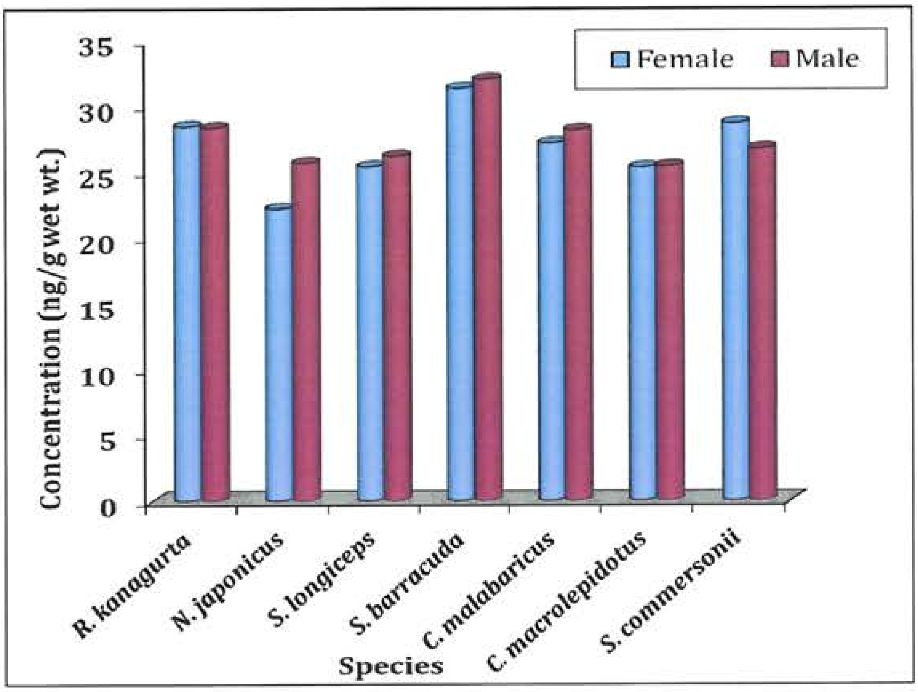


Figure 4.3: Variation in concentration of total PCBs between sex.

4.3. Variation in total PCB concentrations in fishes among various quarters during the study period

Seven species of marine fishes collected between October 2004 and September 2006 which falls into eight Quarters namely Quarter - 1 (October 2004 and December 2004), Quarter - 2 (January 2005 to March 2005), Quarter - 3 (April 2005 and June 2005), Quarter - 4 (July 2005 and September 2005), Quarter - 5 (October 2005 to December 2005), Quarter - 6 (January 2006 and March 2006), Quarter - 7 (April 2006 and June 2006; Quarter - 8: July 2006 and September 2006) were analysed for PCBs based on human consumption and availability in both the locations throughout the year.

During the study period, in Rameshwaram, the maximum concentration of total PCB 40.35 ng/g was recorded in *Sphyraena barracuda* during quarter 8 and the minimum in *Carangoides malabaricus* (17.14 ng/g) during quarter 1 (**Figure 4.4**). Whereas in Cochin, the maximum total PCB contamination was (39.80 ng/g) in *Sardinella longiceps* during quarter 8 and the minimum 16.17 ng/g in both *Cyanoglossus macrolepidotus* (quarter 7) and *Scomberomonus commersonii* (quarter 2) (**Figure 4.5**).

There were significant variations ($p<0.05$) in the pattern of total PCBs observed among eight quarters. In *Nemipterus japonicus* the concentration of total PCBs in quarter 1 was 23.6 ng/g and it decreased to 20.6 ng/g wet wt during quarter 2. However, further increase in concentration was observed in subsequent quarters. Similarly all the seven species tested have more or less similar pattern of accumulation among various quarters. In general, fishes received from Rameshwaram had higher concentration of total PCB during quarter 8 when compared to other quarters.

Rastrelliger *kanagurta* received from Rameshwaram showed uniform pattern of total PCB accumulation during quarter 1, 2, 3 and 4 with the mean concentration ranging between 25.06 ng/g and 25.8 ng/g (**Figure 4.4**). However, unvarying pattern of PCB accumulation were noticed in *Sphyraena barracuda* (28.82 ng/g- quarter 1), (29.78 ng/g- quarter 3 and 35.4 ng/g- quarter 5). While significant variations in total PCBs concentration were observed among *Rastrelliger* *kanagurta, Nemipterus japonicus* and *Carangoides malabaricus* ($p<0.05$), between *Sardinella longiceps* and *Cyanoglossus macrolepidotus* it was not significant ($p>0.05$).

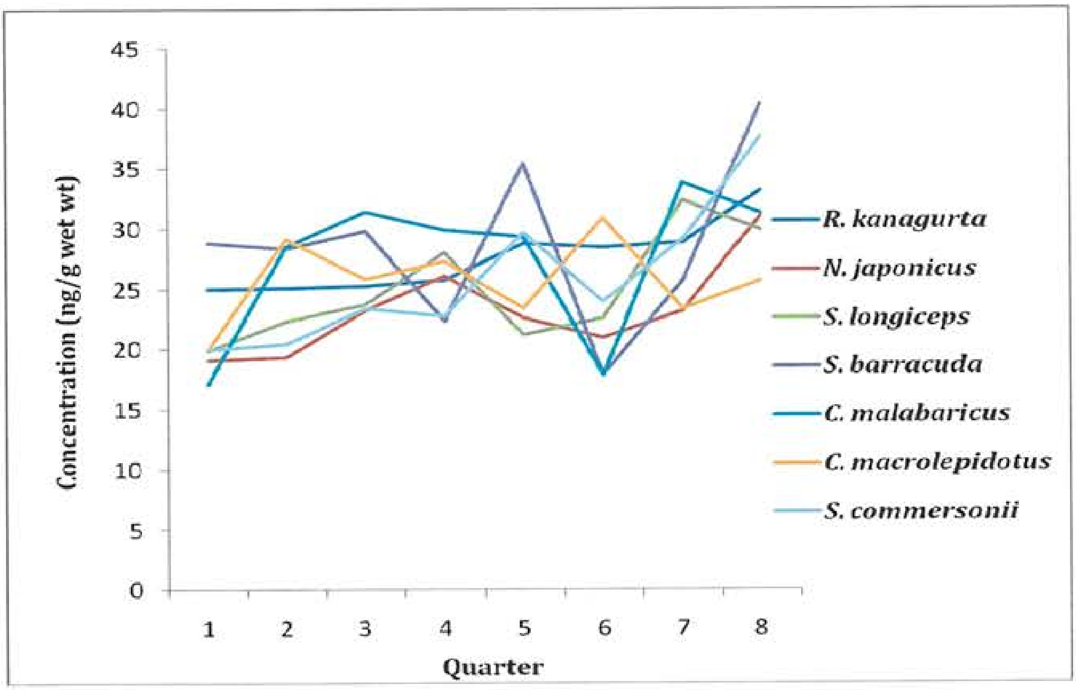


Figure 4.4: Variation in total PCB concentration in fishes of Rameshwaram among various quarters during the study period (Values are in Mean.

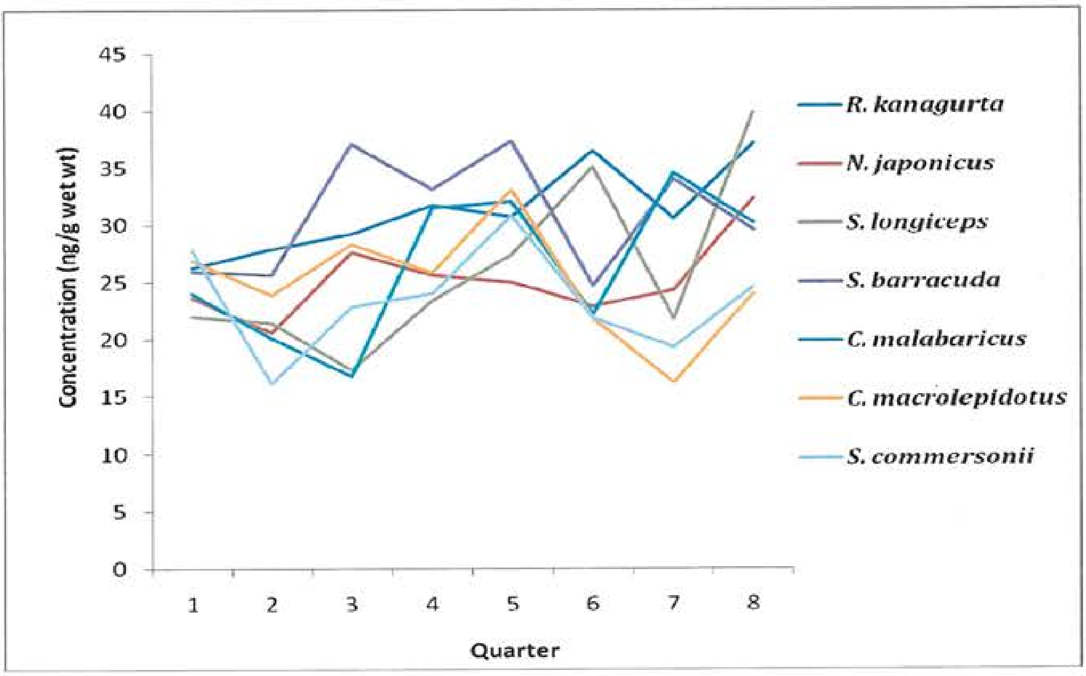


Figure 4.5: Variation in total PCB concentration in fishes of Cochin among various quarters during the study period (Values are in Mean).

4.4. Correlation between size of fishes and total PCB concentration

Pearson's rank correlation was adapted to know the level of correction. The accumulation pattern of PCBs may vary among different size class (length) and food habits of species. Similar observations were reported in many other studies. However, the species presently studied showed negative correlation between size and concentration of total PCBs (**Figure 4.7 a, b, c, d, e, f and g**). Although negative correlation was observed among all the seven species tested individually, the influence of size on concentration was not significant ($p > 0.05$) (**Table 4.2**).

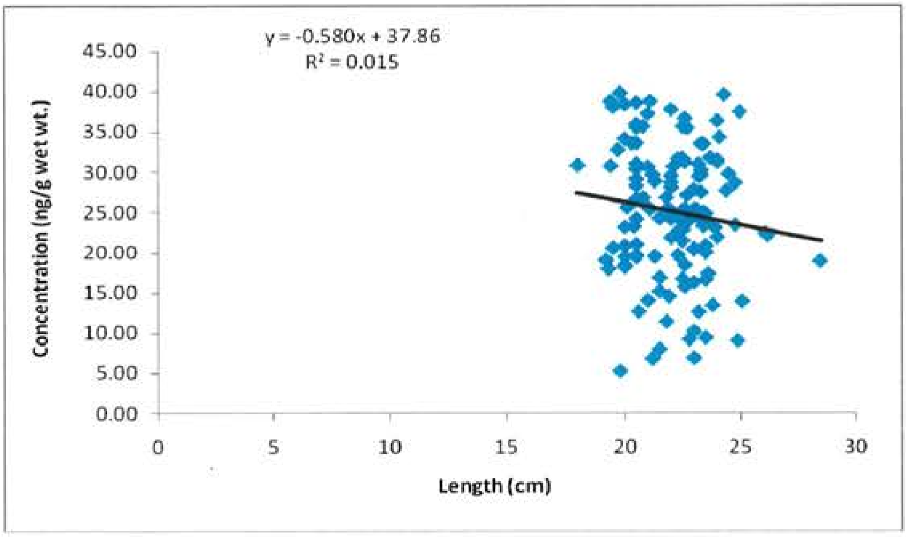


Figure 4.6a: Correlation between size and total PCB concentration in *Rastrelliger kanagurta.*

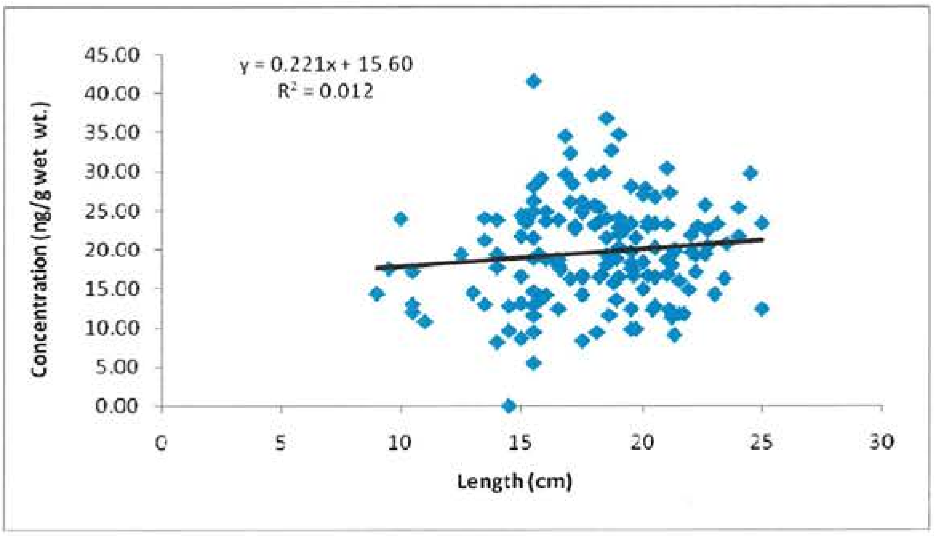


Figure 4.6b: Correlation between size and total PCB concentration in *Nemipterus japonicus.*

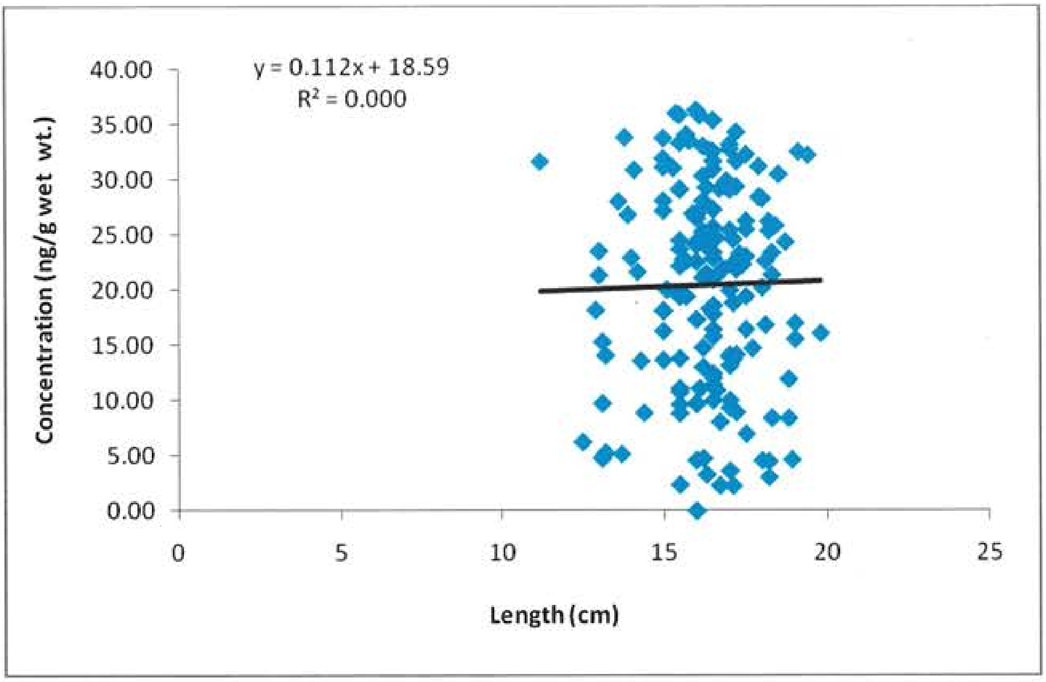


Figure 4.6c: Correlation between size and total PCB concentration in *Sardinella longiceps*.

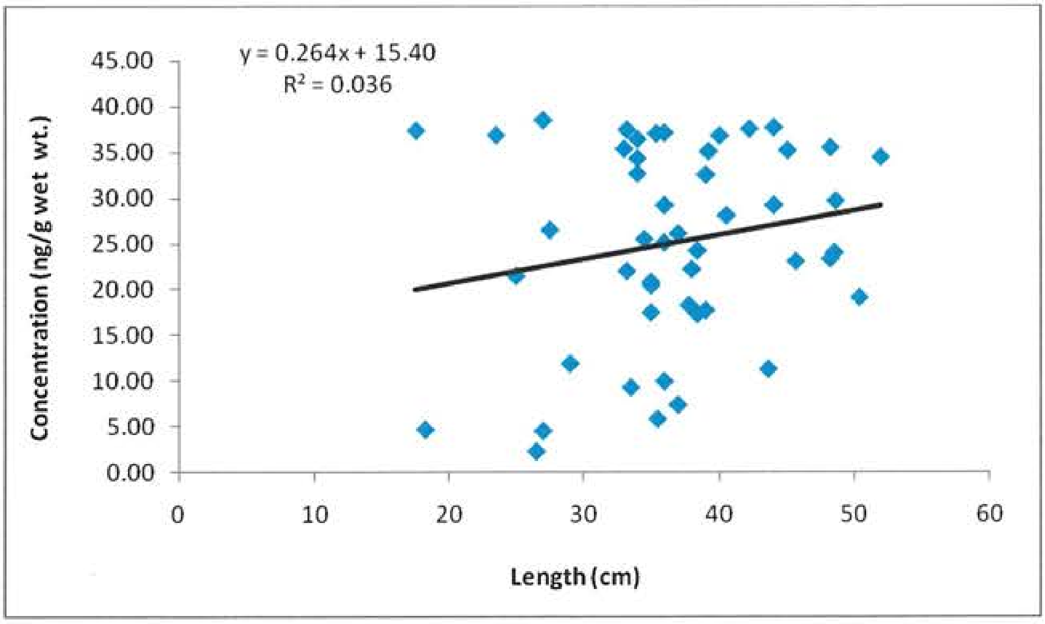


Figure 4.6d: Correlation between size and total PCB concentration in *Sphyraena barracuda*.

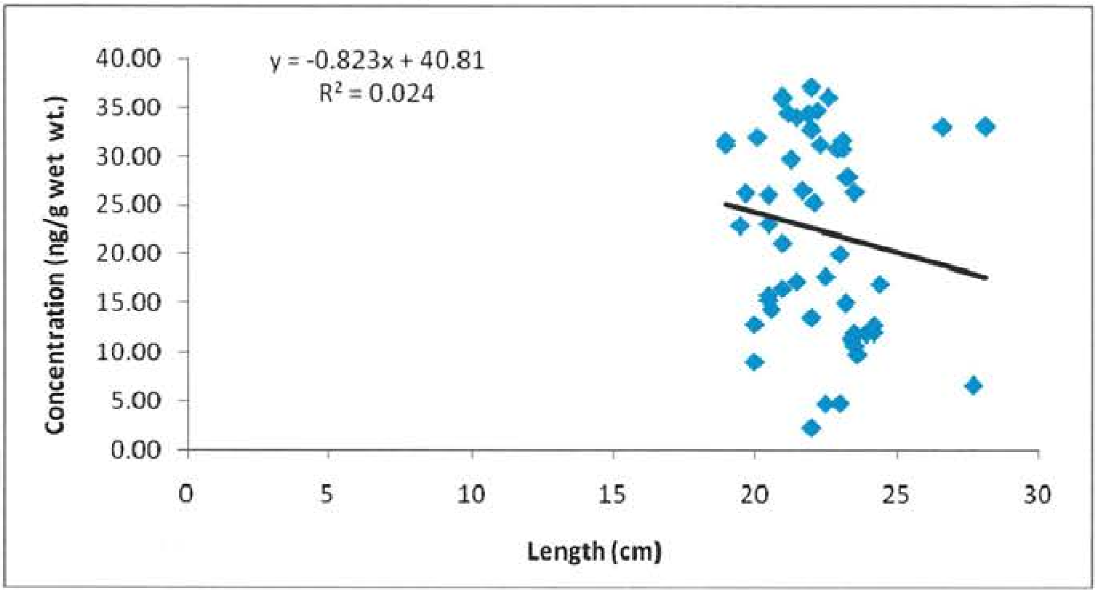


Figure 4.6e: Correlation between size and total PCB concentration in *Carangoides malabaricus*.

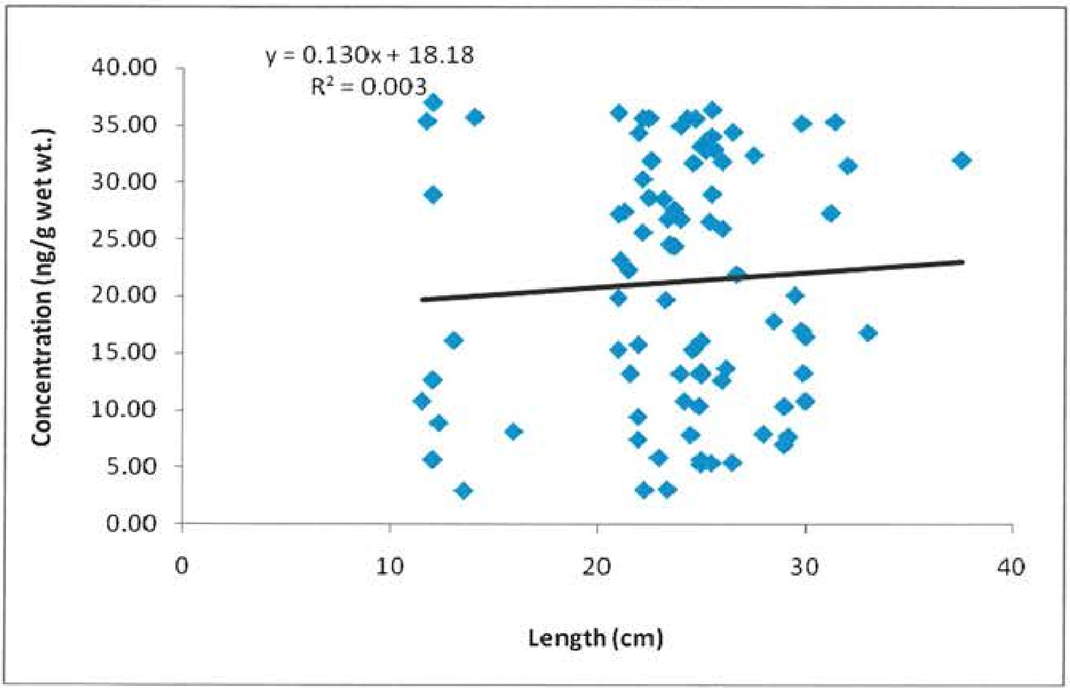


Figure 4.6f: Correlation between size and total PCB concentration in *Cyanoglossus macrolepidotus*.

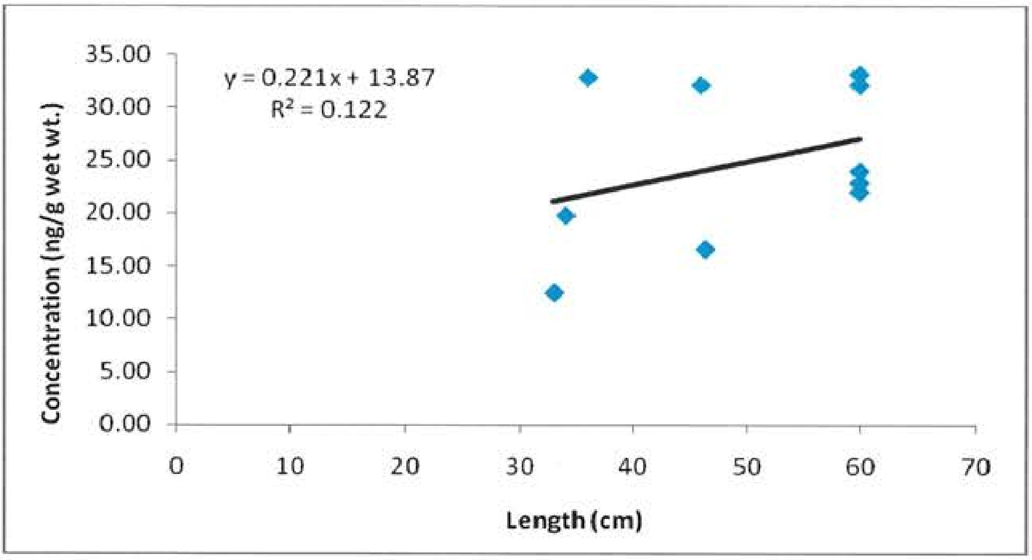


Figure 4.6g: Correlation between size and total PCB concentration in *Scomberomonus commersonii.*

Table 4.2: Correlation between PCB concentrations and body size of fishes studied: coefficient values.

Species	Correlation coefficient value	R^2 value	p value
Rastrelliger kanagurta	Y= -0.5804 x + 37.867	0.015	0.164
Nemipterus japonicus	Y= 0.2216 x + 15.602	0.0121	0.180
Sardinella longiceps	Y= 0.1125 x + 18.595	0.0003	0.818
Sphyraena barracuda	Y= 0.2645 x + 15.401	0.0369	0.186
Carangoides malabaricus	Y= -0.8231 x + 40.811	0.0241	0.959
Cyanoglossus macrolepidotus	Y= 0.1305 x + 18.189	0.0039	0.760
Scomberomonus commersonii	Y= 0.221 x + 13.87	0.1227	0.321

p< 0.05- Significant; p>0.05- Not significant

DISCUSSION

5.1. Selection of fish species

According to 1993 US EPA guidelines, the most important criterion for selecting target fish for contaminant monitoring programs and assessing human health concerns was that the species should be commonly consumed in the study area and of commercial, recreational or subsistence fishing value. Two other criteria of major importance are that the species have the potential to bioaccumulate high concentrations of chemical contaminants and have a wide geographic distribution. In the present study all seven fish species selected satisfied all the three above-referred criteria (Muralidharan et al., 2009).

5.2. PCB Contamination

Thirty two PCB congeners, including six indicator PCBs (PCB 28, 52, 101, 138, 153 and 180) in the muscle tissues of seven species of fishes received from two coastal areas were quantitatively determined. Total PCB concentrations among fish species varied from 23.91 ng/g to 30.15 ng/g. Over 95 % of the fishes studied had levels above the US EPA screening value of 20 ng/g (US EPA, 2000). According to US Food and Drug Administration (FDA) the general population can consume commercially available fish up to 2 mg/kg with no adverse health effect (US FDA, http://www.cfsan.fda.gov/~dms/admehg3.html). Distribution pattern and concentration of toxic contaminants (PCBs) in fish tissues reflect the contamination of the surrounding water and sediment (Bright et al., 1995; Brown et al., 1998; US EPA 2011).

Usage of PCBs reduced in recent years in closed systems such as capacitors and transformers across the world (Bordajandi *et al.,* 2003). The estimates have put the total global production of PCBs in the order of 1.5 million tons (Breivik, 2002). The United States was the single largest producer with over 6,00,000 tons produced between 1930 and 1977. The European region follows with nearly 450,000 tons till 1984. It is unlikely that a full inventory of global PCB production will ever be accurately tallied, as there were factories in Poland, East Germany and

Austria that produced unknown amounts of PCBs (Breivik, 2002). A study by the World Bank in 1996 estimated PCB existence in India at 2000 - 4000 MT (Devanathan *et al.*, 2009). The presence of up to 100 ng/g wet wt of PCBs had been reported in resident piscivorous birds from India (Tanabe *et al.*, 1998) suggesting that concentrations of PCBs reflect the exposure of PCBs in their breeding and feeding grounds within Indian regions.

PCB residues detected in this study reflect the prevalence of the contaminants among various environment of India. Presence of PCBs in Southern Indian water (Tanabe *et al.*, 1982) and Antarctic water (Tanabe *et al.*, 1983) were reported. Wide range of PCBs in water (Kannan *et al.*, 1995), dolphins from Chilika (Kannan *et al.*, 2005) and West Bengal (Karuppiah *et al.*, 2005) were reported in India. Presence of PCBs in the present and earlier studies clearly indicates the occurrence of PCBs in different regions of the Indian Ocean and the ubiquity of such substance in the environment. Sarkar (1994) found varying levels of PCBs (17.6 - 105 ng/g) comprising 31 congeners in zooplankton from the Arabian Sea. According to the authors the sources of PCBs were the coastal industrial installations. It was reported that the East coast of India is much more contaminated than the West coast because of the input of huge amount of organochlorine compounds into the ocean through the river runoff, industrial discharge, as well as agricultural drainage (Sarkar, 1994).

Congener profiles of PCBs in core sediments of Sunderban mangrove wetland and their ecotoxicological significance were well recorded in earlier studies (Minh *et al.*, 2006). Amplified contamination by POPs could be due to dumping sites. Senthilkumar *et al.*, (1999) already reported that penta-Chlorinated Biphenyl (CB) congeners were accumulated predominately in the fishes collected from the Ganges River located in the northern part of India.

5.3. Variation in total PCB concentrations in the fishes collected between Cochin and Rameshwaram coast.

The PCB levels found in muscles tissues of fishes from Cochin are lower than those reported in the fishes from Rameshwaram. Further the levels found in fish muscles from both Cochin and Rameshwaram are lower than those reported by Kannan *et al.*,

(1992) in different species of fishes were obtained from different markets of India (New Delhi, Mumbai, Kolkata, Chennai, Chidambaram and Parangipetti) and from Southeast coast of India (Senthil Kumar *et al.,* 2001). Higher concentrations of chlorinated biphenyls have been reported in fishes, animal fat (goat, lamb and country chicken), liver and blubber of Ganges River dolphins, birds and human fat tissues in southern and northern regions of India reflecting the pollution status of India (Senthil Kumar *et al.,* 2001). On wet weight basis, the concentration of total PCBs reported in the present study (24.5 ng/g) are lower than the levels reported by Senthil Kumar *et al.,* (2001) in Indian Sardine (*Sardinella longiceps*) and Golden Anchovy (*Coillia dussumieri*)] from the Bay of Bengal and sold at Chennai market. These concentrations are lesser than levels reported earlier from Patna (Kannan *et al.,* 1994 and Senthil kumar *et al.,* 1999). In general, studies carried out till-date have shown that the concentration of PCBs in Indian environment is much less than that in developed countries such as the United States and Japan (Kannan *et al.,* 1995).

The maximum concentration of total PCB residues was detected in *Cyanoglossus macrolepidotus* and *Nemipterus japonicus* received from Cochin, while *Sardinella longiceps* and *Rastrelliger kanagurta* received from Rameshwaram showed relatively higher concentration. Total PCB concentration in *Carangoides malabaricus* received from both the locations was more or less the same. The observed spatial variations in total PCB residues although not statistically significant, are due to many factors such as contaminant load in food base, climatic factor like temperature of the water body and annual growth of the fish. Moreover, source of contamination including sewage intrusion and industrial activities around the coastal area influence the accumulation load of contaminants in the fishes (Kannan *et al.,* 1992; Gerstenberger *et al.,* 2002).

The Cochin coastal area does receive effluents from chemical and fish processing industries in addition to the effluents generated by domestic and recreational activities. The effluent load is estimated to be 0.15 million metric tons/ day. Further, the solid waste generated is approximately 2431 tons/day. Port and defence activities also contribute to the contamination load. Rameshwaram, an equally contaminated coastal area receives effluents from industrial establishments such as leather processing, aquaculture, chemicals and automobile.

Approximately, 0.35 million metric tons/day of effluent generated through these industrial establishments are let into the sea (Sharma, 2000). Thus the variation in total residues in the fishes could be mainly contributed by the contaminant load. Busy shipping routes appear to be yet another contaminating source in Indian coasts.

5.4. Variation in total PCB concentration among the fishes included in the study and comparison with earlier studies

Almost all the seven species of fishes included in the study showed similar pattern of PCB accumulation. Total PCB concentrations reported in this study in species, namely *Rastrelliger kanagurta* is concordant with the levels reported in the same species and also prawns collected along the East and West Coast of India (30 ng/g wet wt) (Kannan *et al.*, 1992) and lesser than the concentration reported in fishes of South African Coast (De Kock and Lord *et al.*, 1986).

The concentration of total PCBs in *Sardinella longiceps* reported in the present study is similar to the levels reported in Sardine (24.33 ng/g wet wt.) and Anchovy (20.87 ng/g wet wt.) collected from Catalonia, Spain (Marti - Cid *et al.*, 2007). Although, the variations in PCB concentration among species are not significant, the marginal difference observed among species may be due to the difference in the body fat content, age and feeding habits.

Further, the concentration recorded in the species included in the present study is 10 times higher than the concentration reported in fish (0.2 ng/g, wet wt) and other seafood (2.5 ng/g wet wt.) in China (Nakata *et al.*, 2002) and lower than the levels in the fishes reported by Stefanalli *et al.*, (2004) with 0.9 - 1.7 ppm of PCBs in Adriatic Sea at various locations. Muralidharan *et al.* (2009) had reported high concentration of organochlorine pesticides in 10 species of fishes collected from the same locations. It has been reported that PCBs are present in air, surface water, snow and fishes, namely Lake trout (*Salvelinus hamaycush*) and Rainbow Trout (*Oncorhynchus mykiss*) in the Lake Tahoe basin (Datta *et al.*, 1998).

Yamaguchi *et al.*, (2003) reported the total PCB (mean) concentration 1.83 ng/g in different fishes of Upper Thames catchment area, UK is found to be lower than the presently studied Perch-like fishes namely *Rastrelliger kanagurta* (29.09 ng/g), *Nemipterus japonicus* (24.13 ng/g), *Sphyraena barracuda* (30.15 ng/g), *Carangoides malabaricus* (26.93 ng/g), and *Scomberomorus commersonii* (23.91 ng/g). Gerstenberger *et al.*, (1997a) had reported PCB concentrations (wet weight) in fishes such as Walleye (193.3 ng/g), Pink Salmon (17.3 ng/g), Siscowet Trout (370.3 ng/g), Carp (1,404 ng/g) and Whitefish (154.0 ng/g) collected from Lake Superior, USA. However, the concentrations reported in the present study are lower than the levels reported in the above-referred study.

Guzzella *et al.* (2005) evaluated the presence of PCB residues and other contaminants in the sediments of Hugli estuary, West Bengal and suggested the possible source to be human and industrial activity. The concentration of total PCBs among the species currently studied were in the order of *Scomberomonus commersonii* < *Nemipterus japonicus* < *Sardinella longiceps* < *Cyanoglossus macrolepidotus* < *Carangoides malabaricus* < *Rastrelliger kanagurta* < *Sphyraena barracuda*. The concentration of total PCBs in the present study are higher than the levels reported in edible marine species (2.15 to 132 pg/g) from Adriatic Sea and they reported that Mackerel had higher concentration of PCBs (94.1 - 177 ng/g) (Bayarri *et al.*, 2001) than currently recorded (29.09 ng/g). Concentrations recorded in the present study are several folds lower than the average total PCBs concentrations reported in 12 edible fish species sampled from the Central fish market in Istanbul, Marmara Sea, Turkey (Coelhan *et al.*, 2006).

5.5. Variation in total PCB concentration between sex of different species of fishes collected from Cochin and Rameshwaram coast.

Comparatively, total PCB congeners accumulated higher in male *Nemipterus japonicus* than female (**Figure 4.3**). Variations in PCB concentrations between in fishes are reported to be due to exchange across respiratory surfaces, fugacity -

driven absorption during digestion, egesting and biological differences. For e.g. females lose a substantial portion of their PCB body burden during spawning. Further relative mobility, lipid deposition and metabolic conversion are also the major factors that could explain variation between sex (Morrison *et al.,* 1997; Madenjian *et al.,* 2011). However, sexual differences in PCB concentrations could be inconsistent as reported in the Hudson River suggesting that sexual differences in bioaccumulations could change across ecosystems (Rypel *et al.,* 2007).

If both genders have similar dietary inputs of PCBs, but varying degree of loss, it is obvious that the gender showing greater loss would have lower mean PCB concentrations. In many fishes, females translocate large amounts of body lipid to eggs and some female fishes have been shown to mobilize PCBs from their body into their eggs; this trend is not typically observed in males (Niimi and Oliver, 1983; Sundberg *et al.,* 2005). Alternately, if females consumed greater amounts of prey than males to acquire the metabolic reserves necessary to allow lipid translocation to eggs, they would experience greater dietary exposure to PCBs that could offset or even reverse the trend towards lower PCB concentrations due to spawning losses. In addition, differential habitat utilization by the two sexes could alter this relationship. PCB contamination is known to be localized in many systems and any behaviour that increases time spent within areas of highest concentration or lead to differential consumption of prey from these could lead to higher concentrations in either gender (Zlokovitz and Secor, 1999).

5.6. Variation in body size and total PCB concentration in different species of fishes collected from Cochin and Rameshwaram coast.

In general, negative correlation was observed between the concentration of PCBs and the fish size for many of the species studied presently. However a clear trend was difficult to perceive in all the species. Albaiges *et al.* (1987) had attributed such a concentration decrease in Mediterranean Red Mullet to a possible increase in metabolic activity with age. In the case of female pike, the decrease in pollutants with age was explained by seasonal elimination of lipophilic compounds (Larsson *et al.,* 1992). A reasonable explanation could also be a simple dilution effect produced by the increase of the body weight (Niimi and Oliver, 1983). But, increase in chlorinated hydrocarbons with increase in body size was noted in fishes of Antarctic region (Subramanian *et al.,* 1983). Although the negative correlation was observed among many of the species tested, PCBs effectively do not show significant difference among the size groups, thus suggesting that the concentration reaches equilibrium between uptake and depuration. It is also reported that the substantial bioaccumulation of contaminants is probably due to the level of contamination of the habitat and body fat.

Although, positive correlation between length of the fish and total PCB concentration was observed in the present investigation, it was not significant. Similar correlation was reported in *Anguilla anguilla* collected from Vaccares lagoon, France (Roche *et al.,* 2000). Species specific differences and lipid content of fishes also could influence the accumulation pattern of organochlorine residues among species (Storelli *et al.,* 2007; Szlinder - Richert *et al.,* 2009). Swackhamer and Hites (1988) found that chemical concentrations in lake fish as a function of size were neither similar between species nor consistent among compounds, indicating that physico chemical properties alone do not determine bioaccumulation, but fish characteristics played an equally important role. They reported that, among species, the large size group had the highest concentrations of contaminants, whereas in some species no specific size class consistently had the highest concentrations. Higher levels of PCBs were probably due to bigger size and higher lipid content (WHO, 1993).

5.7. Variation in total PCB concentrations in fishes among different seasons during the study period

The data on levels of PCBs in fishes collected during October 2004 – September 2006 were analyzed to check if there were any temporal trends. For meaningful comparisons, same species collected in all eight quarters were selected. Even though fluctuations were observed in PCB congeners among some of the quarters, similar trend was not observed uniformly in all the species studied. An earlier study (Muralidharan *et al.,* 2009) reported variations in organochlorine pesticide residue concentrations in fishes collected from the same study locations. Further it also reported about 50 % of the species studied to have shown significant variations in total OCs among the seasons. Annual variations in PCB concentration were also reported in fishes of Baltic Sea during 2002 - 2006 (Szlinder - Richert *et al.,* 2009). PCBs were never produced in India. Little information is available about the import status of PCB raw materials and availability of finished products in India. However, the estimated quantity of PCBs used in India is about 2000 - 4000 MT PCBs (Knight, 1996). One of the sources of release of PCBs is scrapping of merchant ships. It is estimated that scrapping of one ship generates about 0.25 - 0.80 MT of PCBs. (Hess *et al.,* 2001).

PCBs containing oils were mostly used in a wide variety of open systems, such as oil paints and exterior emulsions. It is reasonable to assume that this may lead to environmental contamination with PCBs. PCBs have been used in manufacturing of capacitors and transformers. In India, there are number of transformer manufacturers but they do have neither an inventory of the quantity of PCBs being used nor the quantity of PCB- containing equipment. Manufacturers and officials dealing with the transformer oil were not aware about the presence of PCBs in oils. Till-date Central Power Research Institute, Bangalore (CPRI) has identified 2620 tons of pure PCB and 5000 tons of PCB containing equipment in India. Other sources of PCBs may be used oil from abroad, especially from the Middle East region that includes transformer oil containing PCBs mixed in the general pool (CPRI news, 2010).

Present study reveals the continued contamination of the marine environment technical PCB mixtures which are being used in India. Considering an average lifetime of 30 years for large transformers and capacitors, it is presumed that PCB wastes will continue to be generated in the future. In addition, large quantities of PCBs in India are still waiting for disposal. Presence of main congeners namely PCB - 105, PCB - 118 and PCB - 156 (mono - ortho PCBs) and PCB - 101 and PCB - 153 in fishes are indicative of PCB contamination the marine environment and subsequent exposure to fishes. Moreover, half - lives of PCB are approximately 8 to 15 years depending on the specific chemical make - up (Dltri and Kamrin, 1983). According to Wolf *et al.,* (1992) half - lives of persistent PCB congeners, namely PCB 101, 138 and 180 are 8, 40 and 13 years respectively. Although the above estimates may not provide the exact values because only two time points were used for calculations. However, these results imply that exposure of fishes to the above referred PCBs could be continuing and the species might be still at high risk.

Studies explaining temporal variations in PCB residues in fishes in India are very limited. The present study shows a constant increase in the contaminant burden in some of the species, namely, *Sardinella longiceps, Sphyraena barracuda and Carangoides malabaricus.* In addition to this the age of the fish and other physiological factors related to climate change may also contribute to the difference in the contamination pattern among various seasons. Moreover, the study done by Dua *et al.,* (1998) in water bodies (five Lakes of Nainital) of India indicates that atmospheric transport of pollutants, onset of rains, misuse of chemicals in agriculture may contribute to the variations in contamination of the water bodies with respect to seasons. Since there are not many studies available to relate these factors, the discussion on observed variations is restricted. Further, assessment of the seasonal trends is complicated since different factors including physiology of fishes and their feeding habits do not allow any accurate interpretation of the temporal fluctuations observed in contamination (Porte and Albaiges 1993).

Sarkar and Gupta (1987) had reported high levels of organochlorine residues in sediments of the Arabian Sea along the central west coast of India from where also samples for the present study were collected. Hence, it is logical to presume that the contaminants from the sediments ultimately found their way into the marine fishes through the food.

5.8. Tolerance limits or Maximum Residue Levels (MRL) for PCBs

Maximum Residue Level set by the European Union Directive 1999/788 permits a maximum residue levels of 200 ng/g (lipid wt) for the sum of seven main PCB congeners (PCB 28, 52, 101, 118, 138, 153, 180) in edible animals (Li *et al.,* 2008). The total PCBs levels recorded in the present study ranged between 22.65 ng/g and 30.87 ng/g wet wt. Data generated in the present study do not exceed FDA tolerance (action) levels for total PCBs in fish (2 ppm wet wt) (Skinner, 1992) and appear to be within the prescribed limit. WHO (1993) quoted experimentally determined bioconcentration factors (BCFs) in various aquatic species, ranging from 200 to 70,000 or more. In the open ocean, there is bioaccumulation of PCBs in higher trophic levels with an increased proportion of higher chlorinated biphenyls in high - ranking predators. However, in the present study, BCF was not included.

The concentrations of PCBs determined in the present study (22.65 ng/g -30.87 ng/g wet wt) were compared with the tolerance limits set forth by the National Health and Medical Research Council (NHMRC) of the Australian government and Bureau of Chemical Safety, Health and Welfare Canada, 1990). Further, since no tolerance limit has been set for PCBs in foods by the Indian government, the safety limits of the United States Food and Drug Administration (2 mg/kg for fish) ((US FDA) were used.

5.9. Dietary intake and suitability for human consumption

The highly lipophilic PCBs tend to partition into soil and sediment, bioconcentrate from water to aquatic animal, and accumulate in the food chain. Human are high on the food chain; exposure to PCBs and dioxins is mainly from meat, dairy products and fish (Safe, 1994). Bioaccumulations of these persistent contaminants in fishes causing major ill effects in humans have been widely documented all around the world (Cabes *et al.,* 1999; Menone *et al.,* 2000; Vigano *et al.,* 2000). Maternal consumption of PCB contaminated fish in human has been reported to cause reduced birth weight, diminished head and reduced gestational age. PCBs have caused adverse effects on reproduction in a wide range of laboratory animals and decreased survival of offspring (Borrell *et al.,* 2004). In humans, a higher percentage of women who were exposed to PCBs and PCDFs through the consumption of accidentally poisoned cooking oil (the Yucheng accident) reported abnormal menstrual bleeding when compared to unexposed controls (Yu *et al.,* 2000).

On the other hand, menstrual cycle length reductions have been found among women who had consumed more than one meal per month of PCB contaminated fish from the Great Lakes, as well as for women who consumed this fish for seven years or more (Mendola *et al.,* 1997). The consumption of contaminated fat food can be a potential risk for the consumer. To evaluate this risk an attempt has been made to know the possible exposure to human beings through the consumption of contaminated fishes. For this purpose, daily dietary intake of PCB residues were calculated, considering the average body weight of man to be 60 kg, and he consumes 47 g of fish per day (Muralidharan *et al.,* 2009). The estimated daily intake of total PCBs through fish consumption can be considered safe since it contributes to less than the limit of acceptable daily intake (ADI) of total PCBs. Acceptable daily intakes (ADI) have been set only for PCBs at 20 ng/kg body weight/day (WHO 2003). Based on the present study, all the species tested could be considered for safe for human consumption, provided the consumption does not go beyond per week.

PCBs have been detected as a toxic industrial organochlorine compound in human breast milk from India (Tanabe *et al.,* 1990). PCBs have been detected in human adipose tissue and blood from India (Rao and Banerji, 1988, 1989). These observations suggest that PCBs are widespread even in developing countries. The source of PCB intake by Indians was rather unique, where significant contribution was observed from cereals and vegetable oils as well as from dairy products. The dietary intake of PCBs, HCB, and heptachlor in India are relatively low, which might be due to the minimal contamination of the environment by these compounds (Kannan *et al.,* 1992).

In India, the daily intake of PCBs through the consumption of foodstuffs was estimated at 0.86 μg/person, which is apparently lower than those of the developed nations (Kannan *et al.,* 1992). Contaminant concentrations were compared to toxicity thresholds and wildlife toxicity values from the scientific literature to screen for risk. Criteria for inclusion of a toxicity threshold were that the threshold was based on a whole - body fish concentration and was associated with reproductive performance, growth, or survival in fish.

SUMMARY AND CONCLUSION

- ✓ Seven species of marine fishes totalling 732 individuals caught at Cochin and Rameshwaram coasts, and sold in Ukkadam fish market, Coimbatore between October 2004 and October 2006 were analysed for residues of 32 individual congeners, including six indicator PCBs (PCB 28, 52, 101, 138, 153 and 180) with Gas Chromatograph coupled with Mass Selective Detector.

- ✓ Fishes included in the present study are Indian Mackerel (*Rastrelliger kanagurta)*, Japanese Threadfin Bream (*Nemipterus japonicus*), Oil Sardine (*Sardinella longiceps*), Great Barracuda (*Sphyraena barracuda*), Malabar Travelly (*Carangoides malabaricus*), Tongue Sole (*Cyanoglossus macrolepidotus*) and King Seer (*Scomberomonus commersonii*). Selection was based on human consumption (preference), commercial value and availability throughout the year in both the locations.

- ✓ Total PCB concentrations among fish species varied from 23.91 ng/g to 30.15 ng/g. Over 95 % of the fishes studied had levels above the US EPA screening value of 20 ng/g.

- ✓ Variations (ANOVA) in concentrations of total PCB congeners among species, quarters and between locations in the muscle tissue of select species of fishes were tested. Pearson's rank correlation was applied to evaluate the correlation between residue concentrations and the size (length) of the fishes. Student's t-test was adopted to check the variations between sexes.

- ✓ Among the fishes collected from Rameshwaram coast, the highest load of total PCB contamination was observed in *Sphyraena barracuda* (29.64 ± 2.14 ng/g) and the lowest in *Nemipterus japonicus* (22.65 ± 1.99 ng/g). Whereas in Cochin, the maximum residues were recorded in *Rastrelliger kanagurta* (30.88

± 0.97 ng/g) and minimum in *Scomberomonus commersonii* (22.87 ± 1.49 ng/g).

- ✓ Based on ANOVA, while differences in PCB concentration among species were highly significant ($P<0.05$) there was no significant variation between locations ($P>0.05$). Hence the fishes of both the locations (Cochin and Rameshwaram) were considered together for further data analysis.

- ✓ Variation in total PCB concentrations between sexes was not significant ($P>0.05$). Further, pattern of total PCB load was not similar in all the species of fishes analysed. Comparatively higher load of total PCBs was recorded in both male and female *Sphyraena barracuda;* 32.00 ± 1.89 ng/g and 31.32 ± 4.03 ng/g respectively.

- ✓ Among various quarters, in the fishes of Rameshwaram, the maximum load of total PCB (40.35 ng/g) was noticed in *Sphyraena barracuda* (quarter 8) and minimum (17.14 ng/g) in *Carangoides malabaricus* (quarter 1). While in Cochin, the maximum contamination (39.80 ng/g) was in *Sardinella longiceps* during quarter 8 and minimum (16.17 ng/g) was in both *Cyanoglossus macrolepidotus* (quarter 7) and *Scomberomonus commersonii* (quarter 2).

- ✓ Sources of PCB contamination in the fishes are automobile, leather processing, aquaculture and fish processing industries located on the referred coast lines in addition to the busy shipping activities.

- ✓ Even though PCBs were never produced in India, it is estimated that scrapping of one ship generates about 0.25 - 0.80 MT of PCBs. It is to be noted that there are many ship-breaking establishments in the Indian Ports. PCBs containing oils mostly used in transformers and hydraulic fluids are identified as high-risk sources.

- ✓ Magnitude of total PCB contamination among the species currently studied was in the order of *Scomberomonus commersonii* < *Nemipterus japonicus* < *Sardinella longiceps* < *Cyanoglossus macrolepidotus* < *Carangoides malabaricus* < *Rastrelliger kanagurta* < *Sphyraena barracuda.*

- ✓ Studies explaining temporal variations in PCB residues in fishes in India are limited. Present study shows a constant increase through the quarters in contaminant burden in some of the species, namely *Sardinella longiceps, Sphyraena barracuda and Carangoides malabaricus.* Age of the fishes and other physiological factors related to climate change may also contribute to the differences in the contamination pattern among various quarters.

- ✓ PCBs effectively do not show any significant difference among different size classes. Negative correlation was observed in all the species tested.

- ✓ Even though fluctuations were observed in PCB congeners among some of the quarters, similar trend was not observed uniformly for all the species studied.

- ✓ Fish is the primary source of protein through diet to the residents particularly living along the coastal region.
- ✓ Since no tolerance limits have been set for PCBs in fishes by the Indian government, the safety limits set by the US Food and Drug Administration (US FDA) (2 mg/kg for fish) are used.

- ✓ Data generated in the present study do not exceed FDA tolerance levels for total PCBs in any of the species of fish and appear to be within the prescribed limit.

- ✓ Based on the present study, Concentration of total PCBs in the marine fishes sold in Coimbatore market is safe for human consumption and may not cause any adverse health effects to the consumers of Coimbatore. However, it should be reviewed periodically with great concern, as these are hazardous to man as well as to many organisms including, birds.

- ✓ Findings of the present study confirm continued exposure to PCBs through consumption of fish. EPA's peer reviewed cancer reassessment concluded that PCBs are probable human carcinogens. EPA is not alone in its conclusions. The International Agency for Research on Cancer has declared PCBs to be a probable group of carcinogenens to humans. The National Toxicology Program has stated that it is reasonable to conclude that PCBs are carcinogenic in humans. The National Institute for Occupational Safety and Health has determined that PCBs are a potential occupational carcinogen.

- ✓ Although we may expect the environmental residue levels of PCBs to decline in the Indian environment, health concerns are still large in this country.

- ✓ Further the total PCBs levels reported in the present investigation appear to be lesser than the levels reported in many other countries. However, since very little work has been carried out in India, the data generated in the present study could only serve as baseline values. Continued detailed investigations are necessary to understand the situation better.

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