



Assessment of Polychlorinated Biphenyl Congeners in Surface Waters of Langkawi Island, Malaysia

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Abstract

Polychlorinated biphenyl congeners (PCBs) are one of the organic pollutants that were banned in Malaysia because of their toxicity. However, due to their resistant and lipophilic nature, they still detected and posed risks to aquatic animals and human health. The present study is conducted to comprehensively investigate the levels and ecological risks of PCBs in the seawater of Langkawi Island. The unfiltered water samples were collected from five stations around the Geopark and Tourist Island, extracted, and cleaned according to the USEPA method 3510C and 3630C. The PCB congeners, 19 compounds, were analyzed by GC- μ ECD (Agilent). The total concentrations of PCBs in water samples ranged between 188 and 291 ng/L (mean 222 ± 35.6 ng/L). A particularly high concentration of PCBs was detected in Langkawi port station due to its location as the point that receives raw materials as well as chemicals products and cement export. The occurrence of these compounds in Langkawi Island can be attributed mainly to transboundary pollution, wet deposition, and leakage of lubricant oil from boats and/or dumping sites. Compared to the U.S. Environmental Protection Agency (US EPA) guideline, the study results do not pose any health risks to aquatic life, human health, or economic loss.

Keywords: Polychlorinated biphenyls; Water pollution; Langkawi Island; Malaysia.

1. Introduction

Organochlorine compounds such as polychlorinated biphenyls (PCBs) are among the most widespread contaminants in the World. They are considered one of the most persistent organic pollutant groups highlighted by the Stockholm Convention (USEPA, 2009). The hydrophobic nature of these compounds favors them to sorb onto suspended particulate matter and get their way to marine animals through the food chain or deposited in sediments (Moon, et al. 2009). Moreover, the resistance and lipophilic nature of PCB congeners have led to their highly accumulated and amplified through the food web which has been associated with endocrine-disrupting effects and cancer in humans and wildlife (Moon, et al. 2009). PCBs can remain in sediment for many years because of their resistance to degrade and release to the water column by bottom animals or water movements. Highly chlorinated PCBs (with 6-9 chlorine atoms) may be more toxic than PCBs with lower chlorination (USEPA, 2008). PCBs are used in most industrial applications such as lubricating oil, dielectric fluids in transformers and capacitors, stabilizers in paint, polymers, additives in pesticides, and adhesives (Erickson, 1997). Currently, PCB mixtures are still being discharged into different media of the environment from the incineration of chemical wastes, illegal or inappropriate disposal of industrial wastes, and accidental releases.

The Langkawi Archipelago consists of 104 islands, the largest and most exploited of which is Langkawi Island (JPDSM, 2007). Langkawi Island covering an area of 478.5 km², is located in Kedah State, west-north of Peninsular Malaysia in the sea off the Straits of Malacca. The Kedah government declared Langkawi Island as a tourist city in 2005 and officially announced the island as Geopark on the 31st of May 2006 (JPDSM, 2007 and Nasher et al., 2013). In Malaysia, the import of Polychlorinated biphenyls (PCBs) and their products have been banned since 1995. However there are old transformers and capacitors still in use that contain PCB-contaminated oil (Ibrahim, 2007). Monirith et al. (2003) reported that the total PCB congeners in mussel samples

collected from 13 different sites throughout Malaysia ranged between (4 –250) ng/g which were significantly higher when compared with other countries. PCBs have also been detected in high concentrations with a range from 23 to 450 ng/g lipid weight in human breast milk from mothers living in Penang and Kedah states, Malaysia Sudaryanto et al., (2005). The information on the environmental levels of PCB compounds is still lacking in most locations of Malaysia. Therefore, this study aimed to determine the concentrations, distributions, possible sources, and ecological risks of 19 PCB congeners in unfiltered seawater collected from the tourist Langkawi Island.

2. Materials & Methods

2.1. Study area

Figure 1 shows the locations of five sampling stations around the Island including Langkawi Port, Telaga Harbour, Kilim Jetty, Porto Malai Jetty, and Kuah Jetty. These sampling stations were chosen based on their unique activities. The Langkawi Port receives raw materials, and chemical products, and exports cement. Telaga Harbour is a terminal Jetty for sailing boats and yachts, while Kilim Jetty and PortoMalai Jetty are both starting points for marine ecotourism activities including eagle feeding, coastal bat cave exploration, visits to fish farms, and trips to the limestone landscape and mangrove forests. Kuah Jetty is the main terminal for ferries from the mainland, including Kuala Kedah and Penang, as well as Thailand and Singapore (Nasher et al., 2013). The climate of the Island is characterized by dry season, from December to mid-April, and wet or rainy season from May to mid-November. The temperature ranges from 23°C to 35°C (IIOA, 2022). The direction of the wind is generally from Northeast during the Northeast monsoon and from Northwest during the Southwest monsoon. The Island is affected by boundary winds that come from China and Thailand.. Rain deposition increases during the wet season.

2.2. Samples collection

The hydrophobic nature of PCBs causes them to bind to suspended particulate matter in aquatic environments more than remain dissolved. Water samples (1 L), including both dissolved and particulate phases, were collected in precleaned glass bottles from the surface layer. They were then sealed with aluminum foil to avoid photooxidation and kept at pH $\approx$ 2.0 by adding hydrochloric acid (HCl) (12 M) to prevent degradation of the organic pollutants (Moon et al., 2009). The samples were kept at 4°C in ice during transportation and then kept cool at -5°C until analyzed. During sampling water quality parameters temperature, pH, conductivity, and total dissolved solids, were measured in situ, using a surveyor 4a instrument (HACH, USA) (Nasher et al., 2013).

2.3. Chemical analysis

Polychlorinated biphenyl congeners (PCBs) were extracted from one liter of unfiltered water sample according to the USEPA method 3510C (liquid-liquid extraction) (USEPA, 1996) with some modifications which included digital shaker and Ultrasound water- bath for extensive extraction. The clean-up process followed the procedures described in the EPA-method 3630C (USEPA, 1996). Briefly, a 250 mL sample was spiked with 1 mL of surrogate standard 2,4,5,6- tetrachloro-m-xylene (TCMX), decachlorobiphenyl (PCB 209), and a 30mL of dichloromethane (DCM) were added. The sample was agitated in a digital shaker for 6 hrs, then in an Ultrasound water- bath for 1 hr, and then transferred into a separation funnel. The sample was left for 5 min to separate the water from the organic solvent layer. The bottom of the DCM layer containing the organic pollutants was decanted into a 250 mL round-bottomed flask. The extraction was processed and repeated three times (USEPA, 1996). The combined extract was then allowed to mix with granule-activated copper overnight to remove any sulfur contaminants. The extract was passed through a glass column containing 5 g anhydrous Na₂SO₄ (activated at 400 °C for 4hrs before use) to remove any residual water. The extract concentrated to 3 mL using a rotary evaporator. Hexane (10 mL) was then added as an exchange solvent, and the extract was concentrated to 2 mL by rotary evaporator. The extract was passed through a glass column containing 5 g of deactivated silica gel (5% H₂O) and 1 g of anhydrous Na₂SO₄. The PCB congeners fraction was then eluted using 25 mL of pentane and concentrated to 2 mL using a rotary evaporator (USEPA, 1996). Hexane (10 mL) was added as an exchange solvent. It was evaporated down to 2 mL. The extract was finally reduced to 1 mL under a gentle stream of nitrogen gas. All sample extracts were kept in amber glass vials at -5 °C until analyzed within a week. The extract (1 μ L) was injected into Gas Chromatography (Agilent Technologies, USA) equipped with a micro electron capture detector (μ ECD). The total organic carbon (TOC) was determined on precise volume of water samples using a CHNS (O) Analyzer (Thermo) after prior removal of the inorganic carbon using hydrochloric acid (12M).

2.4. Instrumental analysis

Extract (1 μ L) was injected into gas chromatography (Agilent 6890 technologies, USA) equipped with a micro electron capture detector (μ ECD) and a fused silica DB-5MS capillary column (30m $\times$ 0.25mm i.d.) with the film thickness of 0.25 μ m (Thermo Fisher, USA). High purity N₂ (99.9%) was used as a carrier gas, makeup gas, and purge gas at flow rates of 1.0, 45, and 30.0 mL/min, respectively. The flow rates for the μ ECD were 450 mL/min and 45 mL/min for air and hydrogen, respectively. The gas chromatograph was operated in splitless mode and separation was conducted with the oven temperature. The temperature program was 80 °C (hold 3 min), increased by 20 °C/min to 180 °C (hold 0 min), and increased by 3 °C /min to 280 °C (hold 25 min).

2.5. Quality assurance and quality control QA/QC.

Quality control and quality assurance were applied to all data. Replicate samples were analyzed for each station to calculate the

precision of measurements. The method blank containing the solvents and surrogate internal standard underwent analysis to evaluate the contamination of solvents, reagents, and the glassware that was used. The accuracy of the analytical procedure was examined through the recovery of spiked water after subtraction from the unspiked water. A known amount of the 19 PCB congeners was spiked into a deionized water sample and left for a few hours before being similarly extracted and analyzed. The surrogate internal standard (2,4,5,6- tetrachloro-m-xylene (TCMX) and decachlorobiphenyl (PCB 209)) were used throughout the analytical procedure to monitor the losses and contamination during the sample extraction and instrumental analysis (USEPA, 1996). The average recovery of the 19 PCB congeners ranged from 73 to 96% with relative standard deviation (RSD) values being mainly less than 10%, which meets the acceptance criteria of the method (EPA-method 3630C), thereby indicating that the method adopted from EPA is acceptable for the analysis of PCBs in the unfiltered seawater samples.

2.6. Data analysis

The External standard calibration comprising 19 PCB congeners was used to identify and quantify, each component peak in the sample chromatogram which corresponds to the standard compounds used for calibration purposes. Linear relationships were obtained with correlation coefficients (r) from the linear regression of $r = (0.98 - 0.99)$ for 19 PCB congeners. Regression equations obtained an r value of ≥ 0.99 are acceptable according to the Environmental Protection Agency. Agilent chemstation software was used to obtain the chromatogram and for data calculations. Pearson correlation analysis was used to test the relationship between PCBs and water parameters (e.g. TOC), and PCBs across the studied station using SPSS version 15.0 for Windows. Statistical significance was defined as $p \leq 0.05$ (p-value at the 95% confidence level).

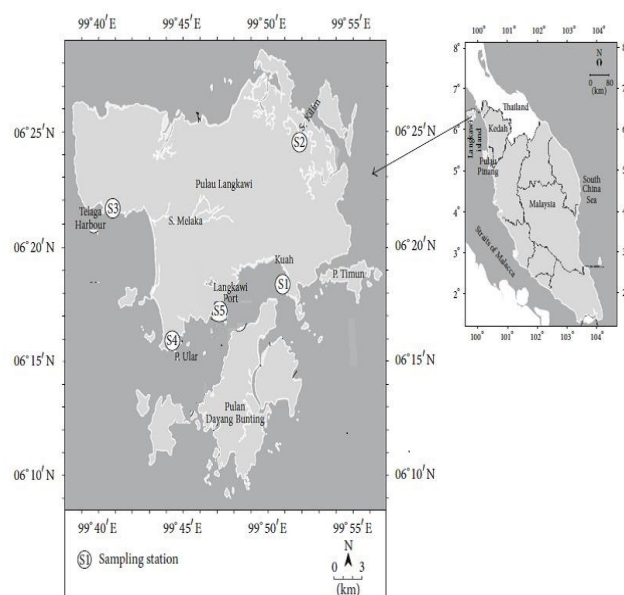


Figure 1. The locations of sampling stations (S1 to S5) around Langkawi Island, Malaysia.

2.7. Ecological risk assessment

To assess the probability that PCB concentrations in Langkawi Island may cause adverse effects to aquatic life, or not. The results were compared with the US-EPA guideline which is known as the safe level for the water of aquatic life and human beings. The concentrations below the US-EPA guidelines suggest that no adverse effects on aquatic life. The concentrations above the limit suggest adverse effects would happen frequently on the aquatic life (ATSDR, 2014).

Table 1. Physico-chemical parameters (mean $\pm$ SD) of water at sampling stations in Langkawi Island.

Sampling stations (code)	D* (m)	Conductivity (μ S $\cdot$ m ⁻¹)	TDS (mg L ⁻¹)	pH	Temperature (°C)	TOC %
Kuah Jetty (S1)	3	47.59 $\pm$ 0.25	50.12 $\pm$ 0.18	7.91 $\pm$ 0.12	28.80 $\pm$ 0.36	0.38
Kilim Jetty (S2)	1	48.61 $\pm$ 0.34	51.37 $\pm$ 0.31	7.45 $\pm$ 0.04	28.57 $\pm$ 0.23	0.03
Telaga Harbour (S3)	3	47.85 $\pm$ 0.12	50.57 $\pm$ 0.10	8.13 $\pm$ 0.01	28.67 $\pm$ 0.32	0.02
Porto malai Jetty (S4)	3	47.68 $\pm$ 0.47	50.27 $\pm$ 0.57	8.24 $\pm$ 0.09	28.60 $\pm$ 0.17	0.07
Langkawi port (S5)	3	47.88 $\pm$ 0.22	50.46 $\pm$ 0.22	8.24 $\pm$ 0.02	29.20 $\pm$ 0.26	0.01

*Depth, TDS: Total dissolved solid, , TOC: Total organic carbon.

3.Results and Discussion

Table 1 shows the measurements of water quality parameters that include conductivity, pH, total dissolved solids, temperature, and total organic carbons. The measurements were in agreement with the Malaysia water quality standard. Figure 2. shows the concentrations of a total of 19 polychlorinated biphenyl congeners (PCBs) in the unfiltered surface water of the five sampling stations around Langkawi Island. The concentrations ranged from 188 $\pm$ 5.6 ng/L at Porto Malai Jetty to 291 $\pm$ 62.2 ng/L at Langkawi port with the mean concentration 222 $\pm$ 35.6 ng/L. The highest concentration of total PCB congeners detected at Langkawi port was due to the port activities received; raw materials and chemical products, and exporting the cement. The persistent and hydrophobic nature of PCBs coupled with their high affinity for suspended particulates, released from the bottom sediment layer and the deposition during rainfall season is known as possible PCBs inputs, the same PCBs inputs were concluded by Chevreuil and Granier (1985) and Yuko Ogata et al. 2009. While the total concentrations of other stations including Kilim Jetty, Telaga Harbor, Kuah Ferry, and Porto Malai Jetty were extremely similar to 220 $\pm$ 12.2, 208 $\pm$ 23.8, 202 $\pm$ 29.3, 188 $\pm$ 5.6 ng/L respectively. The similarity in the total concentrations of PCBs in the different stations reveals the same sources and/or pollutant activities. The wet deposition during the wet season that is characterized by heavy rainfall is considered one of the important PCB sources in tropical areas as indicated by Wurl and Obbard 2005, especially when the PCBs are carried by Northeast winds from neighboring regions such as China and Thailand. For example, Thailand as shown in the study done by Cheevaporn et al. (2005), has high levels of PCBs in urban and industrial areas which resulted in the high use of transformers, capacitors, and waste draining from industrial activities and the power plant. Moreover, the waste disposal of electrical transformers, oil spillage, and sewage from ships may lead to a widespread distribution of PCBs in the marine environment of Southeast Asia (Wurl and Obbard, 2005).

There is no significant correlation between total PCB congeners and total organic carbons (TOC%) ($r^2 = 0.02$, $p < 0.05$) which indicate separated primary sources or differing transport processes. There is a good correlation between total PCB congeners concentration and sampling locations ($r^2 = 0.60$) that reflect the same possible sources or transport processes. Additionally, there is no significant correlation between total PCB congeners and water quality parameters; pH, conductivity and total dissolved solid.

However, with temperature, the results revealed a good significant correlation which reflects how the water temperature affected these PCB compounds.

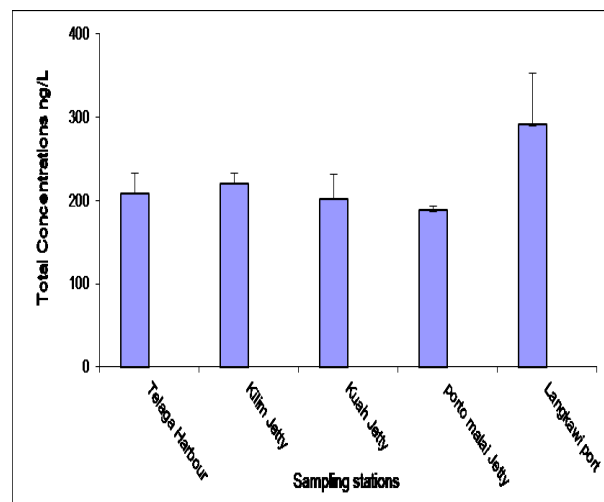


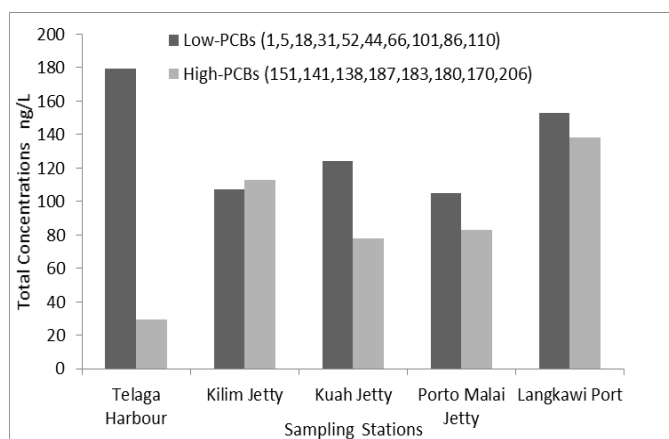
Figure 2. The total concentrations (ng/L) of PCBs for sampling stations of Langkawi Island.

Table 2 shows that the total PCB congeners concentrations in the water of Langkawi Island were approximately lower by 1 to 4 orders of magnitude than the water investigated by other studies in Otamiri River, Imo State, USA (Eze, et al., 2023), the coastal seawater of Rijeka Bay-Northern Adriatic (Nena and picer 1992), the Minjiang River Estuary, Southeast China (Zhang et al., 2003) and Niger Delta, Udu River, Nigeria (Iniaghe and Kpomah, 2022). However, the total PCB congeners in the Langkawi water were approximately 1 to 10 orders of magnitude higher than those detected in other countries, such as the marine water in the South Baltic Sea (Sapota, G. 2004), Xiamen Harbour, China (Zhou, et al., 2000), Singapore (Basheer et al., 2003), The Venice Lagoon-(Manodori.,2006) (Table 2). One of the reasons for the high concentration in this study is that water samples in this study analyzed the PCBs in the dissolved and particulates together. While some other studies covered only the dissolved water which made the comparison unclear. Another reason is due to the number of PCB congeners where some studies included 7 PCB congeners and others 12 or 13 PCB while in this study a number of 19 PCB congeners were covered.

Table 2. Comparison of the total PCB concentrations (ng/L) in water of this study with those collected from the existing studies.

Site	No of congeners	Concentrations rang (ng/L)	References
Minjiang Estuary	22	203.9 - 2473	Zhang et al., 2003
River Otamiri, Imo State, USA	40	622.87-935.84	Eze, et al., 2023
Rijeka Bay-Northern Adriatic	00	28.0 – 597	Nena and picer 1992
South Baltic Sea	7	0.24 - 2.07	Sapota, G. 2004
Xiamen Harbour, China	12	0.1 - 1.7	Zhou, et al., 2000
Singapore	40	0. 04 – 61.76	Basheer et al., 2003
The Venice Lagoon,	13	1.2 - 10.5	Manodori.,2006
Niger Delta, Udu River, Nigeria	28	20-1860	Iniahe PO, Kpomah, 2022
Bay of Bengal coast of Bangladesh	209	32.1-199.4	H. Al-Mamun et al., 2019
Langkawi Island, Malaysia	19	188 - 291	Present Study

Figure 3. shows the total concentrations of the low-chlorinated PCBs and the high-chlorinated PCBs for sampling stations of Langkawi Island. The lower-chlorinated PCBs were found to be higher than that of the higher-chlorinated PCBs for most samples. This suggests that the low-PCBs predominated over high-PCBs was attributed to low water solubility and preference of the high-PCBs for a particulate phase or/and sediment (Eze, et al., 2023 and Baqar et al., 2017). Studies have reported that PCBs decline as the number of chlorine atoms increases (Iwegbue et al., 2020). As a result, high-PCBs have a low degree of mobility and they are deposited close to the source of emissions (Al-Wabel et al 2016); and they are re-suspended to the water column from the sediment. Low-PCBs, on the other hand, are more prevalent due to their high mobility, ability to accumulate in the atmosphere, ability to be effectively scavenged by precipitation, and ability to be transported by water currents and runoff from the Island to the aquatic environment (Eze, et al., 2023 & Gao et al., 2013). Besides, the presence of high low-PCBs concentrations indicates that dechlorination processes, long-distance migration (e.g.Thailand), and atmospheric deposition from dumping sites are improbable to be the only factors influencing their distribution in Langkawi Island. Therefore, the distribution of both low-PCBs and high-PCBs in the Island is significantly influenced by local and boundary sources.

**Figure 3.** The total concentrations (ng/L) of the low-chlorinated PCBs and the high-chlorinated PCBs for sampling stations of Langkawi Island.

PCB congeners have been classified by the United States Environmental Protection Agency (ATSDR, 2014) as probable human carcinogens of medium carcinogenic hazard (Group B2), which probably pose carcinogenic effects to the marine life. For ecological risk assessment, the study results were compared to the US EPA guideline (ATSDR, 2014). The maximum contaminant level (MCL) of PCBs in drinking water should not exceed 0.0005

mg/L (500 ng/L). The total PCB congeners in all water samples (188 -291 ng/L) were below the MCL, suggesting that the potential risks to aquatic life and human health for the Langkawi population and tourists are low.

4. CONCLUSION

The total concentrations of 19 PCB congeners in the unfiltered water of Langkawi Island were measured. The highest concentration was found, in Langkawi port, due to the port activities. The predominance of low-PCBs over high-PCBs in most locations was attributed to low water solubility and their high degree of mobility and ability to be transported by winds and water currents from neighboring regions. The results show that there are no health risks to marine resources, tourists, and local people of the Island according to the US EPA guidelines. However, future studies on PCBs and other pollutants in the different media of the Island environment are deemed recommended to monitor and assess the health of Langkawi Island. That will be a good early warning for the Langkawi Development Authority (LADA) and the Langkawi Municipal to develop strategies for protecting the Island by mitigating or avoiding the sources of risks.

Data Availability

No data were used to support this study.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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