DISTRIBUTIONS OF ORGANOCHLORINE PESTICIDES IN SEDIMENT AND AQUATIC BIOTA: BENZENEHEXACHLORIDES (BHCS)

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ABSTRACT

The usage of organochlorine pesticides (OCPs), which is also known as persistent organic pollutants (POPs), is either prohibited or restricted. However, a number of studies on sediments and aquatic biotas including fish and shellfish revealed that most of the compounds for instance benzenehexachlorides (BHCs) group are still present in the environment. Aquatic biotas can be used as bioindicator to monitor the concentrations of BHCs in the aquatic systems such as rivers and oceans. At the same time, sediments can also be used to investigate the existence of BHCs in the environment. Sediments serve as a home for benthic biota such as worms and clams, which are commonly consumed by fish and benthic feeders. OCPs are easily absorbed on suspended particulate organic matter due to their high octanol–water partition coefficient (Kow) which accumulated in bed sediments, which makes the sediments as potential contamination source.

Keywords: *Organochlorine Pesticides (OCPs), Benzenehexachlorides (BHCs), Shellfish, Sediment*

1. INTRODUCTION

High concentrations of organochlorine pesticides (OCPs) have been a major environmental issue (Taiwo, 2019). According to Ali et al. (2019), OCPs were extensively used to increase the production of agricultural crops. OCPs were the most widely used pesticides around the world before 1970s in both agricultural and industrial sectors as highlighted in Wang et al. (2019). Zhu et al. (2019) reported that the presence of OCPs in sediments may exist due to desorption of residual OCPs from soil and sediment contaminated from previous fumigation activities.

During flash floods, rainwater could have washed off contaminated soil and cause residual OCPs to enter the river water, and finally settled in the sediments. Pesticides run-off will lead to contamination of sediments and biota; dysfunction of ecological system in water system due to the loss of top predators resulting from growth inhabitations and reproductive failure and public health impacts from eating contaminated fish and shellfish (Thangaraj, 2016). The activities in the long term, may affect nature, where OCPs for instance can remain in the soil for up to 30 years (Zhang et al., 2018). For that reason, this paper reviews the distribution of benzenehexachlorides (BHCs) which is one of the OCPs group in sediments and aquatic biotas samples from all over the world.

2. BENZENEHEXACHLORIDES (BHCS) GROUP

Benzenehexachloride or BHC was first discovered in 1825. It is made by chlorinating benzene, which results in a product made up of several isomers, i.e. molecules containing the same number of atoms but differ in the internal arrangements of those atoms. BHC has four isomers, named after the Greek letters; alpha, beta, gamma and delta as shown in Figure 1 (Shinggu et al., 2015). After much laboratory work in isolating and identifying these isomers, only the gamma isomer has insecticidal properties.

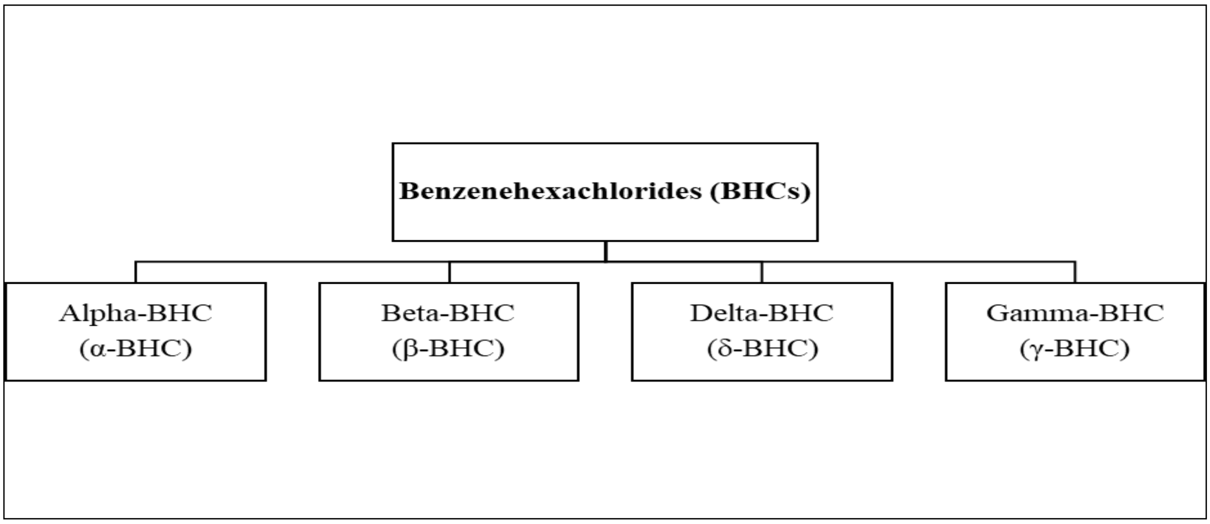

Figure 1: Subgroup of BHCs

The gamma isomer makes up almost 15 percent in the technical material and used for a limited type of application, such as seed treatment in industrialized countries. Gamma isomer or more popular as lindane, is more toxic than the alpha or delta isomers from fifty to several thousand times (Vijgen et al., 2019). Lindane has been extensively used worldwide for the control of agricultural pest. The low aqueous solubility and chlorinated nature of this compound contribute to its persistence and resistance to degradation by microorganisms.

The effects of lindane on insects and mammals apparently look like those of DDT (Yang et al., 2019). It is also a lipophilic compound and therefore tends to accumulate and concentrate in the fat of animals and humans (Harit, 2019). There have been reports on the occurrence of lindane residues in soil, water, air, plants, agricultural products, animals, food, microbial environment, and humans (Boudh, & Singh, 2019). In addition, due to its widespread use and persistence, lindane is a common, ubiquitous pollutant found in the biosphere (Tyutikov, 2018).

2.1 BHCs in Sediments

BHCs have penetrated waterway in many parts of the world and found to accumulate and persist in bed sediments as mentioned by Smyth and Grieco, 2018. In general, sediment serves as a home for benthic biota such as insects and clams, which are commonly consumed by fish. It can also be an important sink for BHCs which has been used in the past and at present (Duodu et al., 2016). Referring to Ogbeide et al., (2015) and Sakan et al. (2017), the sediment could also be a potential source for BHCS contamination through a series of biogeochemical processes. Concentrations of BHCs in sediment may be affected by several factors such as agricultural surface run-off, groundwater movement, atmospheric environment and leaching process.

From another point of view, although sediment does not relate directly with the degree of aquatic pollution, it helps in reconstructing historical inputs of anthropogenic pollutants as mentioned by Kumar and Mukherji, (2018). Due to its long range transportation, Olutona, et al. (2016) found the presence of a widespread contamination of BHCs in sediment, even though the pesticide activities do not take place in the sampling area. To make it worse, BHCs easily absorb particulate matter and finally accumulate in sediment because of their hydrophobic nature and low solubility in water (Bhardwaj & Kapley, 2015). In conclusion, due to its composition, geographic distribution and potential environment, the significance of sediment is important and needs to be discussed especially in explaining the high concentration of BHCs (MateoSagasta, & Tare, 2016).

It is believed that the first study on BHCs concentrations in Malaysia was done by Iwata et al. (1993) in sediments from Pantai Jeram, Selangor. In the study, BHCs ranged from <0.01 ng/g to 0.11 ng/g. Total concentration of BHCs isomers ranged from ranged from 0.002 to 59.17 ng/g was determined in the sediment samples from Cameron Highlands, Pahang, Malaysia. The ratio of BHCs composition specified a clear historical practice and new contributions of these pesticides in the agriculture area. Gamma isomer was the most dominant compound detected in the sediment samples compared to alpha, beta, gamma and delta isomers of BHCs. The existence of BHCs in sediments is possibly the product of past contamination, discharges in waters, agricultural run-off and atmospheric deposit. Some seasonal differences in the concentration level of BHCs were noted due to the dilution and degradation factors in the sampling locations (Saadati et al., 2012).

Saad (2013) reported that the concentrations of BHCs in sediment samples ranged from 162.00 to 195.60 ng/g dry wt. The sediment samples were taken from Paka River and Lembah Sitrus, in Dungun Terengganu. In the study, the BHCs group was dominated by β-HCH isomer.

The OCPs originated from adjacent agriculture and aquaculture activities nearby. The use of pesticides in farming and aquaculture has commonly been skyrocketing due to increasing population and demand for extra agrochemicals.

A study on OCPs distribution and concentration in sediment was done at northeast coastal area in Sabah (Khamarudin, 2013). Six sampling stations was involved including Pulau Tigabu, Pulau Musa, Pulau Malawi, Pulau Jambongan and Sulu Sulawesi Sea station. Three BHCs isomers including alpha, beta and delta were detected ranging from 0.38 to 21.01 ng/g. Beta-BHC was dominated in all sediment samples from all sampling stations. The existence of these compounds in the sampling stations was related to the location of the stations. As the coastal area, these sites were become dumping area for domestic, agricultural and industrial wastes. Increasing of population made the situation worst. In addition, the OCPs contaminants were suspected to come from inefficient wastewater treatment plants, town run-off and poor waste disposal management along the coastal area. In coastal sediments the concentration of BHCs where OCPs contamination was caused by aquaculture cages and shipping activities near the fishing village.

Abdullah et al. (2015) detected OCPs levels in sediment. The values were ranging from 0.41 to 82.16 ng/g of dry weight. Two isomers of BHCs compounds namely gamma (γ-HCH) and beta (β-HCH) were detected in the sediments respectively. The γ-HCH compound was found in the vegetable plantation zones. On the other hand, β-HCH was identified in the downstream locations of the Telom and Bertam Rivers in Cameron Highland, Pahang, Malaysia. According to the authors, run-off during rainfall brought the OCPs pollutants from agricultural areas and villages into the river.

In another study, three BHCs isomers including α-HCH, β-HCH and γ-HCH were identified in the soils used for vegetables farming from Cameron Highland, Malaysia. The γ-HCH isomers was detected as the highest concentration compared to other isomers in the soils The OCPs concentrations in the soils were ranged from 13.7 to 44.1 µg/kg (Farina et al., 2016). Cameron Highland, Pahang is known as the important producer of vegetables to Malaysia. For that reason, frequent pesticides used were observed in this study area.

The BHCs isomers, α-HCH and γ-HCH were the most frequent identified in the soil samples from paddy field area in Ledang, Johor, Malaysia. The concentrations of 0.182 and 0.183 µg/kg were detected, respectively. The isomeric ratio of BHCs compounds calculated were higher than the parent compound, β-BHC >1 which revealed the point that the existence of BHC isomer is related to elderly use. Concentrations of BCH isomer were established to be in the order of γ-BHC > α-BHC > δ-BHC > β-BHC (Nabhan et al., 2018).

Osman and Khalik (2018) found all BHCs isomers in the sediment samples from ten samples of lowland paddy field cultivation area in Machang, Kelantan, Malaysia. The concentrations were ranged from 0.02 to 7.34 µg/kg. The α-BHC was the most highly detected isomers in all samples followed by β-BHC, γ-BHC and δ-BHC. The OCPs especially BHCs were still detected in agricultural area in Malaysia.

In ASEAN countries, Wurl et al. (2007) revealed the presence of BHCs (3.30 ng/g - 46.20 ng/g) along Singapore coast. The concentrations were caused by rapid industrial development and busy shipping traffic along the coast. In sediments from Bueng Boraphet wetland, Thailand, Chaiyarat et al. (2015)

reported the presence of total BHCs of 87.24 ng/g. The high levels of total BHCs detected in the sediments were endorsed by α-BHC, β-BHC and γ-BHC. The high concentration of total BHCs in sediments were mostly indicated their current banned use as agro-pesticides or herbicides in farming area at Bueng Boraphet wetland. However, one of the isomer BHCs namely δ-BHC was not detected in the sediments from the sampling area.

In the case of Indonesia, sediment samples were collected from Rawa Pening Lake, Central Java. The concentrations of BHCs were mostly attributed by γ-BHC isomer with 0.009 mg/kg. The appearance of OCPs compound specifically BHCs was based on the persistency of this pesticides in the environment. The residues of OCPs may persist and accumulate in the sediment even there were no current usage of OCPs by farmers in this area. However, we cannot rule out the possibility that this pesticides are still used illegally by farmers (Isworo et al., 2015).

Total BHCs of 7.82 ng/g and 6.94 ng/g were detected in sediment samples from CauBay River and KieuKy, Vietnam respectively. The contamination of BHCs were conceived from pesticide usage with regards to crop protection and vector control. These compounds were transferred in the atmosphere from the extensive agricultural zones (Toan, 2015).

In China, Zhonghua et al. (2016) reported the concentration of BHCs (BDL to 113.0 ng/g) in sediment samples from Yangtze River and Huaihe River. The δ-BHC compound was recorded the highest concentration in all samples. In India, Kumar et al. (2018) reported contamination of BHCs (0.01- 2.54 μg/kg) in sediments from Central India. The environmental and human health risk of these toxic compound were observed in the study. Almost hundred sediment samples were extracted by using ultra sonication. The samples were cleaned-up with silica gel. The BHCs compound were detected using gas chromatography electron capture detector (GC-ECD).

The BHCs isomers (α-BHC, β-BHC and γ-HCH), were detected in all sediment samples from Nairobi River, Kenya (Ndunda et al., 2018). The β-HCH was the highest detected in all sampling stations. This probably due to its high environmental stability and low volatility characteristics. Other than that, α-BHC and γ-BHC isomers be able to transform into β-HCH. As a result, β-BHC become dominant isomer in all sites.

In similarities, some OCPs are still being used in rapid developing countries like China and India, mainly for agricultural and industrial purposes, due to their low cost and flexibility, even after the restriction and ban based on the Stockholm convention. Table 1 tabulates BHCs concentrations in sediment samples collected from different Asia countries in the previous studies.

Table 1: BHCs in Sediment Samples from Different Countries

Sampling locations	BHCs concentrations	References
Pantai Jeram, Selangor, Malaysia	<0.01 - 0.11 ng/g	Iwata et al. (1993)
Cameron Highlands, Pahang, Malaysia	0.002 - 59.17 ng/g	Saadati et al. (2012)
Paka River, Dungun, Terengganu, Malaysia	1.62 - 195.60 ng/g	Saad (2013)
Northeast coastal, Sabah	0.38 - 21.01 ng/g	Khamarudin (2013)
Cameron Highland, Pahang, Malaysia	0.41 - 82.16 ng/g	Abdullah et al. (2015)
Cameron Highland, Pahang, Malaysia	13.7 - 44.1 μg/kg	Farina et al. (2016)
Ledang, Johor, Malaysia	0.98 - 3.60 μg/kg	Nabhan et al. (2018)
Machang, Kelantan, Malaysia	0.02 - 7.34 μg/kg	Osman and Khalik (2018)
Singapore Coast	3.30 - 46.2 ng/g	Wurl et al. (2007)

Bueng Boraphet wetland, Thailand	87.24 ng/g	Chaiyarat et al. (2015)
Rawa Pening Lake, Central Java, Indonesia	0.009 mg/kg	Isworo et al. (2015)
CauBay River and KieuKy, Vietnam	6.94 - 7.82 ng/g	Toan, (2015)
Yangtze River and Huaihe River, China	BDL-113.0 ng/g	Zhonghua et al. (2016)
Central India, India	0.01- 2.54 µg/kg	Kumar et al. (2018)
Nairobi River, Kenya	0.01 - 41.9 µg kg^{-1}	Ndunda et al. (2018)

2.2 BHCs in Fish and Shellfish

Pigot (2017) reported that fish and shellfish are essential source of edible food for human and other animals. They contain high protein with low calories that enhance human health. Besides that, fish and shellfish are main source of organic compound namely omega-3 fatty acids. According to Calder, (2015) these fatty acid helps to improve human health with decreasing heart illness among the adults and promote good vision and brain growth for babies as well. As a result, eating pesticides contaminated fish and shellfish by human are the main route source in human and other animals' body systems. Ogbeide et al. (2015) in their studies confirmed that there are positives relationship between pesticides contents in aquatic biotas with human and other animals. Another study by Ali, (2019) also reported that humans are exposed with toxicants for instance pesticides by consumed fish and fish based products such as fish cakes and salted fish. For that reason, concentrations data of BHCs in fish are very important to assess the existence of this harmful pesticides in our food source (Fair et al. 2018).

Fish and shellfish are choosen as bioindicator in monitoring BHCs in our ecosystem because of some reasons including their habitat, their response against the toxicants and the exposure of the pollutant (Elia et al., 2007). If we compare to humans and other animals, fish and shellfish tend to accumulate the pesticides in their body directly from the water where they live. Besides that, they also get contaminated by their food intake (Davodi et al., 2011). The ability of pesticides to bioaccumulate and increase their concentration (biomagnified) in aquatic animals including fish and shellfish also create anxiety to human being (Maurya et al., 2016). The persistency of BHCs in the environment is also a main concern to us (Miranda et al., 2008).

The earliest study was done by Jothy et al. (1983). They reported low concentration of total BHCs in fish and shellfish from the west coast of Peninsular Malaysia ranging from 0.001 mg/kg to 0.01 mg/kg. In 2013, a study by Saad reported concentration of BHCs group in catfish (*Arius maculatus* and *Clarias batrachus*). The concentrations were ranged from 0.90 to 506.85 ng/g. The fish samples were collected from Paka River and stream at Lembah Sitrus, Dungun, Terengganu. The β-BHC was dominated in all fish samples. High concentrations of total BHCs probably caused by high activities of agriculture at that sites that use BHCs to protect their plants (vegetables, oil palm trees, rubber trees and honey orange trees) from pests.

Total BHCs with concentration of 12.00 ng/g was detected in green mussel samples (*Perna viridis*) from Singapore coastal water (Monirith et al., 2003). High concentration of BHCs in this mussel species were due to the feeding pattern as this biotas are benthic feeders and are exposed to the contamination in the aquatic habitat. Besides that, the existence of BHCs in the coastal water of Singapore probably due to the extensive usage of these compounds (BHCs) in many areas including, industries, pharmacies and domestic used (Zhang et al., 2015).

Lower concentrations of total BHCs were found in samples from Indonesia. The BHCs concentrations in various fish samples collected were ranged from 0.78 ng/g to 4.20 ng/g (Sudaryanto et al., 2007). The distributions of BHCs in Indonesia waters were probably caused by the run off of these compounds from extensive agricultures sectors in that country. Hamilton et al. (2016) described that fish that inhabit those areas were exposed to the contaminant and bioaccumulate the toxicants in their body systems. However, lower concentration was detected possibly as result of dilution factor caused by rainfall or the feeding pattern of the fish samples and the habitat occupy by the fish.

Three BHCs isomers (α, β and γ) were identified in fish samples specifically silver barb (*Barbonymus gonionotus*), climbing perch (*Anabas testudineus*), snakehead (*Channa striatus*), river barb (*Cyclocheilichthys enoplos*), mud carp (*Cirrhinus microlepis*) and three-spot gourami (*Trichogaster trichopterus*). The concentration of BHCs were ranged from 0.10 to 1.85 ng/g. All fish samples were caught from paddy field area in Bueng Boraphet wetland, Thailand (Chaiyarat et al., 2015). From the result, the study exhibited that fishes in the rice paddy areas in Bueng Boraphet wetland were contaminated by BHCs compounds.

The BHCs compounds were ranged from 2.84 to106.11 ng/g in various fish species collected along coastal fisheries of China (Pan et al., 2016). The concentration of BHCs were contributed by recent usage of lindane and old technical BHCs compounds. High concentration of total BHCs was found in fish possibly due to high body fat content especially during spawning seasons. The BHCs were tend to attach onto the fat tissues (Kim et al., 2015).

In another case, BHCs compounds were detected in fish samples collected from Indus River, Pakistan. The concentrations were ranged from 0.17 to 22.7 ng/g (Robinson et al., 2016). All isomers were detected in the fish tissues. The α-BHC was dominated in the fish muscle in all samples. The δ-BHC isomer was the least detected compound in the fish. This was due to the most of the technical grade of BHCs compound used in this agriculture area comprised highest percentage of α-BHC.

Bhuvaneshwari and Rajendran, (2012) reported that BHCs concentration at 0.82 to 3.5 ng/g were detected in five fish species (*Etroplus suratensis*, *Oreochromis mossambicus*, *Liza parsia*, *Channa striatus* and *Silurus wynaadensis*). The fish samples were taken from River Cauvery and Veeranam Lake in India. Two isomers of BHCs (α and β) were the most extensively detected in this study. From the result, the extensive distribution of α-BHC in the fish samples probably due to degradation of γ-BHC to α-BHC by microorganisms inhibiting in soil and sediments. On the other side, β-BHC is extremely persistent in the environment (Sineli et al., 2016).

The contamination of BHCs in aquatic biotas were also identified in other countries. A study by Nguyen et al. (2018) detected total BHCs with concentration at 5 to 7159 µg/kg. Fish and mollusks samples were taken from Soai Rap and Long Tau estuary of the Sai Gon - Dong Nai river system in Vietnam. The predominant isomer in this study was β-BHC. It was detected in all fish and mollusks samples. From the result, this study shows high accumulation levels of BHCs compound in biota samples.

High concentration of BHCs were detected (471 to 1570 ng/g) in two fish species *Oreochromis mossambicus* (Mozambique tilapia) and *Clarias gariepinus* (African sharptooth catfish). The fish samples were collected from iSimangaliso Wetland Park in South Africa. As one of the OCPs members, BHCs possess the persistent and bioaccumulation features. That's mean, these compounds can be are easily attached and remain in the fat tissues of non-target including humans and animals. The concentrations of these pesticides in the organisms become higher from one level to another level in the food webs due to the biomagnification characteristics (Xia et al., 2015). Table 2 presents previous studies of BHCs in fish and shellfish samples from different countries.

Table 2: BHCs in Fish and Shellfish from Different Countries

Sampling locations	BHCs concentrations	References
West coast of Peninsular Malaysia	0.001 - 0.01 mg/kg	Jothy et al. (1983)
Paka River and Lembah Sitrus, Dungun, Terengganu	0.90 - 506.85 ng/g	Saad (2013)
Singapore	12.00 ng/g	Monirith et al. (2003)
Indonesia	0.78 - 4.20 ng/g	Sudaryanto et al. (2007)
Bueng Boraphet wetland, Thailand	0.10 - 1.85 ng/g	Chaiyarat et al. (2015)

Coastal Fisheries, China	2.84 -106.11 ng/g	Pan et al. (2016)
Indus River, Pakistan	0.17 - 22.7 ng/g	Robinson et al. (2016)
River Cauvery and Veeranam Lake, India	0.82 - 3.5 ng/g	Bhuvaneshwari, & Rajendran, (2012)
iSimangaliso Wetland Park, South Africa	471-1570 ng/g	Buah-Kwofie et al. (2018)

3. CONCLUSION

Studies on distributions of BHCs from various sampling sites in different countries, may give an idea on the past and present usage of these harmful compounds. It will then be detailed out based on the sources of pollution, where surroundings of sampling stations are the main point to be discussed. In addition, varieties in geographical condition for each sampling station may demonstrate different patterns of pollution, based on the compounds used. Other than that, this review paper provides previous studies information on BHCs concentrations in aquatic biotas and sediments for further references.

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