

Environmental Monitoring Assessment of Organochlorine Insecticides in Palakkad District, Kerala, India

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Abstract: The establishment of organochlorine insecticides (OCIs) exhibited remarkable benefits on various sectors such as agricultural, commercial, pharmaceutical, household usage, active ingredient as preservatives in fruits and vegetables etc. Nevertheless, the use of these synthetics increased day by day, perceived harmful effects worldwide. The present study focused on the current trends in the OCI levels in selected soils of Palakkad district, Kerala. Soil samples were collected during the month of September, 2014 (Wet Season) and February, 2015 (Dry Season) and OCIs namely, α -BHC, β -BHC, γ -BHC, heptachlor, aldrin, heptachlor epoxide (B), 4,4'-DDE, dieldrin, endrin, 2,4'-DDD, 4,4'-DDD, 2,4'-DDT, 4,4'-DDT, α -endosulfan and β -endosulfan, has been monitored in order to assess the caused risk assessment by using GC-ECD. Results revealed that the Σ OCIs in soils were ranged from 16.79 – 123.27 ng g⁻¹ along the stations throughout the sampling period. Dominant contaminants frequently observed were endrin, heptachlor derivatives, aldrin, BHC derivatives and dieldrin, while other analyzed OCIs including DDT derivatives and endosulphan were found below detection levels (BDL). These observations highlight the gradual shift in the application of synthetic towards traditional practices of organic farming in the area. However, the occurrence of trace amounts of OCIs in the realm could be designated as slight recent application.

Keywords: Organochlorine insecticides (OCIs), contaminants, monitoring, environment, soil.

1. INTRODUCTION

Environmental monitoring is the assessment of quality of environs by estimating the concentration, features, trends etc. of indicating parameters. The primary objectives of such programs are to signify the detection and early warning of the contaminant's level of specific site under investigation. One such study could apply on the toxicity assessment of human made persistent pesticides specifically, OCIs. OCIs are a group of synthetically manufactured chemical compounds widely and generally range of applied in agricultural, medicinal and industrial fields. They possess beneficial achievements in various field of activities like controlling vectors of diseases, pests or non-target organisms which harm the effective production and their yield. Benzenehexachloride and its derivatives, heptachlor, aldrin, heptachlor epoxide (B), dieldrin, endrin, DDT derivatives, endosulfan etc. are the representative compounds of these chlorinated hydrocarbons.

Owing to their persistent nature, these compounds remain in the soil niche for a long period of time after its application in different environmental segments. Since they are fat soluble in character, they are readily absorbed in to the body fat of the target insect and thereby threatening the proper functioning of the internal organs of the species. OCIs directly act on both central and peripheral nervous system of the target organism by destructing the mechanism of Na/K pump for providing potential gradient in the nerve cell membrane functioning for generating the necessary energy. These toxic compounds are absorbed immediately after consuming into the alimentary tract or articulately absorbed through skin of the target insects and arthropods. This chemical compound is represented as endocrine disrupting insecticides (Pleština, 2003; Nair and Sujatha, 2012; Victoria et al., 2013). Both in developed/developing and under developed countries, the usages of these xenobiotics are different with respect to their utility. In India the use of insecticides is about 80 %, while comparing worldwide it is about 29.5 %, regards to an enormous levels of consumption (De et al., 2014; Devi et al., 2017).

Apart from their beneficial characteristics, as they are well known for their endocrine disrupting nature, inhibitors of acetyl cholinesterase and potential carcinogen to humans, etc. (Simonich and Hites, 1995; Kamrin, 1997; Hall and Hall, 1999; FAOUN, 2002; Aktar et al., 2009). These water insoluble compounds find their way by transferring and translocating from one place to another through land leaching and runoff and they persist for decades in the environment devoid of degradation. Despite of the fact that, the extensive use of these multipurpose insecticides, leads to their accumulation in the soil system, groundwater column, and finally ends up in the ocean, persist for years in the deep ocean waters, thereby causing toxic to the benthic communities (Reichenberger et al., 2007). Subsequently, these activities facilitate their exposure to higher organisms, aquatic organisms and human beings, triggering nerve related health concerns (Paul and Balasubramaniam, 1997). These toxic pollutants enter in to the life cycle of living organism through a chain of actions transferring them from parental body to fetuses through placenta or by lactation. Hence, the harmful effects will carry generations to generations, resulting to produce a community of handicapped society, causing permanent neurobehavioral impairment (Metcalf, 1997; Paul and Balasubramaniam, 1997).

Toxicity of these class of persistent insecticides and its harmful effect on humanity and non-target organisms led to the prohibition of their production, sale and application/usage in several countries (USEPA, 2009). However, these toxic insecticides were detected in certain parts of the world (Manirakiza et al., 2002; Wabel et al., 2011; Sruthi et al., 2017; Gopalan and Chenicherry, 2018). Therefore, these findings suggest the way that the frequent investigations were essential for assessing the soil quality thereby recommending remedial strategies. The current research endeavor is such an environment monitoring assessment of selected soils of Palakkad district, Kerala. Recently, traces of toxic chemicals such as endrin, 2, 4'-DDD, aldrin, γ -BHC, etc. were detected from Palakkad soils (Gopalan and Chenicherry, 2018). Hence this research theme provides an outlook on the contamination levels in the study area under consideration thereby serving the environmentalists, to gather information to tackle an effective measure for regulate and control over the harmful impacts. Consequently, the objective is to determine the current trends in the residual levels of OCIs: α -BHC, β -BHC, γ -BHC, heptachlor, aldrin, heptachlor epoxide (B), 4,4'-DDE, dieldrin, endrin, 2,4'-DDD, 4,4'-DDD, 2,4'-DDT, 4,4'-DDT, α -endosulfan and β -endosulfan and their possible sources.

2. MATERIALS AND METHODS

2.1 Soil Sampling Collection

Palakkad district is prominent in its agricultural activities and known as the granary of Kerala. It lies between 10°20' north latitude and 11°14' and 70°02' and 76°34' east longitudes (Figure 1). The district experiences a dry climate, exhibits the characteristic of cracking during summer and develops temperature extended to record as 41 °C, moreover periodically changing humid climate prevailing in the low land and midland of this district, could reduce the adsorption potential of their laterite rich soils. A total of six prominent locations were selected viz., C I, C II, C III, K I, K II, K III and these representative soil samples were collected in clean brown glass bottles during the month September (2014) and February (2015). Thereafter, they were transferred to the laboratory and kept at -20 °C till analysis. Prior to the analysis, soil samples were freeze dried and grinded to fine powder by agate mortar.

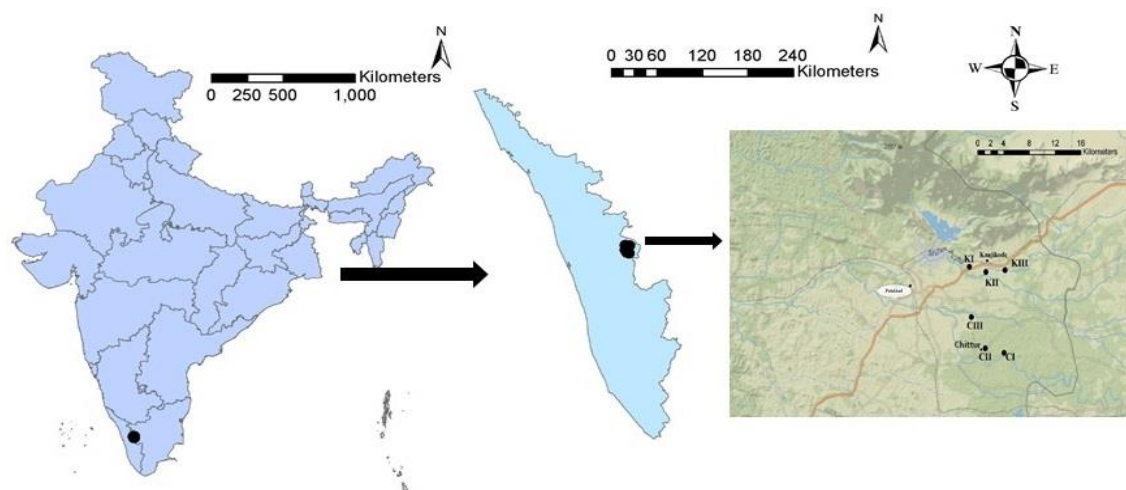


Fig 1: Study area showing the sampling location of Palakkad district

2.2 Analysis of the soil samples

All the chemicals and reagents such as OCIs standards used in the present work were purchased from EPA (USA) and Supelco (USA). About 5 g of the soil samples were accurately weighed and extracted twice with hexane: acetone (1:1) mixture. Later, cleanup procedure was performed by their elution through Florisil column (60 cm x 22mm i.d.) with hexane: acetone (1:1). The extract was concentrated to about 5-6 ml using rotary evaporator at 50 – 60 °C for further analysis. The determination of OCIs were performed via gas chromatograph (GC - model 7890A, Agilent, Waldbronn, Germany) equipped with a split/splitless injector, an electron capture detector (ECD). The separation was performed on a fused silica capillary column: 30 m x 0.320 mm i.d, HP-35 capillary column with 0.5 µm film (35% - (phenyl)-methyl polysiloxane phase). The injector port temperature was maintained at 250 °C, detector temperature at 350 °C. The GC oven temperature was programmed from 110 °C at 5 °C min⁻¹ to 190 °C and this was held for another 2 min, then to 280 °C at a heating rate of 15 °C min⁻¹. This temperature was held constant for 10 min. Nitrogen was used as the carrier gas at a flow rate of 1.0 mL/min; split ratio 1:50.

2.3 Quality Assurance

All samples were extracted and analyzed in triplicate and were subjected to strict quality control procedures. A procedural blank and a spiked sample were run in each set of ten samples to nullify the errors due to contamination of glasswares. The mean recoveries ranging from 85 to 109% and the mean recovery of surrogate standards was 90 ± 5% in the extraction method. The detection limit for all the OCIs ranged from 0.01 to 0.08 ng g⁻¹ respectively.

3. RESULTS AND DISCUSSIONS

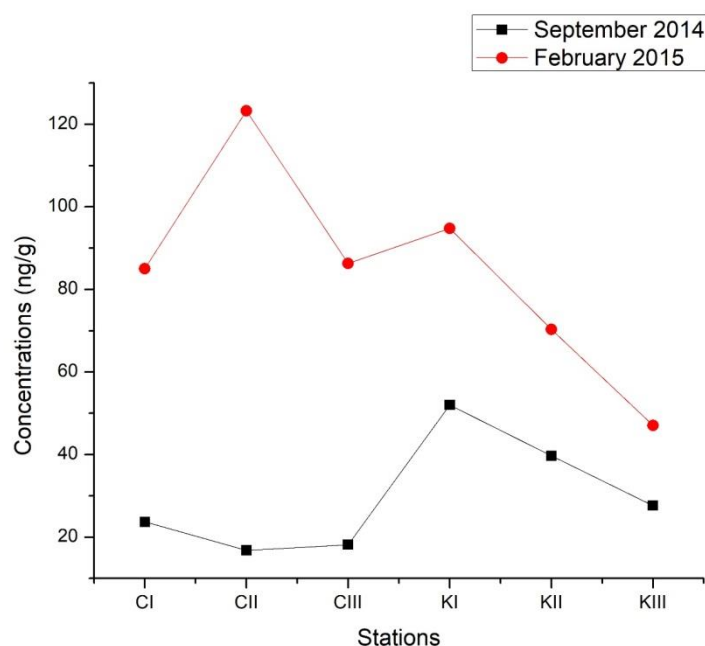


Fig 2: Concentration of Σ OCIs (ng g⁻¹) in the surface soil of Palakkad district during September 2014 and February 2015

OCI residues in the prominent locations of Palakkad district, Kerala, during September, 2014 (wet season) and February, 2015 (dry season) were summarized in Table I & II. Most of the analyzed OCIs including, α -BHC, β -BHC, γ -BHC, heptachlor, aldrin, heptachlor epoxide (B), dieldrin, endrin were present in the sampling station during both periods, except DDT derivatives and endosulphan. The Σ OCIs in the soil samples were ranged from 16.79 – 123.27 ng g⁻¹ (Fig 2). The overall concentrations of OCIs during wet season were comparatively lower than the dry season. The highest values were observed at the station C II (123.27 ng g⁻¹) during wet season. The levels of OCIs in the study region found to higher in the dry season than the wet season which could be washed away along with rainwater and possibly translocated into nearby water column. The present information regarding OCI residues in the study area reveals a gradual decrease in the concentration on comparing with the data set available (Gopalan and Chenicherry, 2018). This could be due to their reduced application into the system and the change in cultivation procedures by preferring organic farming technologies.

BHC derivatives (α -BHC, β -BHC, γ -BHC) were detected in all the samples throughout the sampling period September 2014 and February 2015 with a range from $0.84 \pm 0.2 - 24.94 \pm 2.1 \text{ ng g}^{-1}$ and $5.38 \pm 2.1 - 31.16 \pm 1.2 \text{ ng g}^{-1}$ respectively. Similar observations were recorded in earlier studies in soils of Palakkad, BDL - $15.9 \pm 3.4 \text{ ng g}^{-1}$ (Gopalan and Chenicherry, 2018). Likewise, soils of various environs showcased different trends such as in Kuttanad agro ecosystem, Korba (Central India), Shanghai, UK, Korea, European high altitude mountains and rural soil of Hong Kong were ranged from BDL - 8.91 (Sruthi et al., 2017), $0.9 - 20 \mu\text{g kg}^{-1}$ (Kumar et al., 2014), ND to 10.38 ng g^{-1} , $2.4 \pm 0.72 \text{ ng g}^{-1}$ (Jiang et al., 2009; Nakata et al., 2005), $0.1-10 \text{ ng g}^{-1}$ (Meijer et al., 2001), $<3 \text{ ng g}^{-1}$ (Kim and Smith, 2001), $0.08-0.49 \text{ ng g}^{-1}$ (Grimalt et al., 2004), $6.19 \pm 1.31 \text{ ng g}^{-1}$ (Zhang et al., 2006). Furthermore, the levels of BHC derivatives in various parts of India were depicted in Table III. Among the analyzed BHC derivatives, β -BHC was dominant isomer observed and these could be due to current application in to the soil (Andreu and Picó 2004; Stewart and Chisholm 1971). Additionally, β -BHC is the most stable form, which are resistant to microbial as well as photolytic degradation among the other derivatives. Thereby, they adsorb in the soil along with the soil organic matter moieties and persist (Kalbitz et al. 1997; Mackay et al. 1997). According to Willett and his teammates (1998) hypothesis, α and γ -BHC isomers generally convert to β -BHC could also triggers the accumulation of β isomer in the soil. Contrary to the reports of Ramesh et al., (1991) and Takeoka et al., (1991), in the present research work, the concentration of BHC isomers were observed in higher levels in dry season than the wet season.

TABLE I: Concentration of OCIs (ng g^{-1}) in the surface soil of Palakkad district during September 2014

Sampling No.	Stations	α -BHC	β -BHC	γ -BHC	HEPTACHLOR	ALDRIN	HEPTACHLOR EPOXIDE (B)	DIELDRIN	ENDRIN	4,4'-DDE	2,4'-DDD	4,4'-DDD	2,4'-DDT	4,4'-DDT	α -ENDO	β -ENDO
September 2014	CI	0.94 ± 0.1	7.14 ± 2.1	11.04 ± 1.1	2.03 ± 0.1	2.54 ± 0.1	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
	CII	0.84 ± 0.2	10.02 ± 1.2	2.75 ± 2.1	1.6 0.1	1.58 0.2	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
	CIII	0.85 ± 0.1	6.83 ± 2.1	1.57 ± 3.1	0.98 ± 0.3	3.32 0.12	4.61 ± 2.1	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
	KI	3.51 ± 1.1	24.94 ± 2.1	8.7 0.2	5.69 ± 1.1	1.46 0.1	BDL	7.71 ± 1.1	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
	KII	1.38 ± 1.2	16.46 ± 2.2	6.6 1.2	5.34 ± 1.2	2.33 1.1	7.55 ± 1.2	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
	KIII	0.94 ± 0.1	7.16 ± 2.1	6.89 ± 1.2	4.29 ± 0.1	2.15 1.1	6.2 1.2	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL

Table II: Concentration of OCIs (ng g^{-1}) in the surface soil of Palakkad district during February 2015

Sampling No.	Stations	α -BHC	β -BHC	γ -BHC	HEPTACHLOR	ALDRIN	HEPTACHLOR EPOXIDE (B)	DIELDRIN	ENDRIN	4,4'-DDE	2,4'-DDD	4,4'-DDD	2,4'-DDT	4,4'-DDT	α -ENDO	β -ENDO
February 2015	CI	5.38 ± 2.1	31.11 ± 2.1	8.53 2.1	BDL	15.28 ± 1.1	24.73 ± 2.4	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
	CII	6.38 1.2	31.15 ± 4.5	8.53 1.1	BDL	15.27 ± 1.4	24.66 ± 2.1	BDL	37.28 2.1	BDL	BDL	BDL	BDL	BDL	BDL	BDL
	CIII	6.52 0.1	31.13 ± 3.1	8.53 2.1	BDL	15.27 ± 1.2	24.82 ± 3.1	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
	KI	6.72 0.2	31.12 ± 1.2	9.01 2.4	7.88 1.2	15.27 ± 4.1	24.79 ± 1.4	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
	KII	6.83 1.1	31.15 ± 2.1	9.12 2.1	7.96 2.4	15.28 ± 2.6	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
	KIII	6.83 2.2	31.16 ± 1.2	9.08 2.1	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL

Besides BHCs, the presence of metabolites of chlordane i.e., heptachlor and heptachlor epoxide (B) were detected in the study area which lies within the range from BDL - $24.82 \pm 3.1 \text{ ng g}^{-1}$. During the dry season, higher levels of heptachlor epoxide (B) were observed. The occurrence of higher levels of heptachlor epoxide (B) possibly due to the degradation product of heptachlor (Keith, 1997). Previous studies in the study region also detected residual levels of these toxic compounds and indicating their prolonged application (Gopalan and Chenicherry, 2018). These substances readily adsorb into the animal tissues and could enter into the food web further triggering health issues (USEPA, 1986; Singh et al., 2007).

Table III: BHC derivatives (ng g^{-1}) in various parts of India

Location and Type of Soil	BHC Derivatives	References
Palakkad Surface soil	BDL - 15.9 ± 3.4	Gopalan and Chenicherry, 2018
Kuttanad Agro ecosystem	BDL - 8.91	Sruthi et al., 2017
Hyderabad Surface soil	75.89	Kata et al., 2015
Korba Residential soil	0.9 - 16	Kumar et al., 2014
Assam Forest soil	0.007 - 0.025	Devi et al., 2013
Tripura Forest soil	0.003 - 0.049	Devi et al., 2013
Manipur Forest soil	ND* - 1.15	Devi et al., 2013
Kurukshetra Surface soil	0.6 - 8.5	Kumar et al., 2013
Keoladeo National Park Sediment	9039.9	Bhadouria et al., 2012
NCR Delhi Surface soil	34.96	Kumar et al., 2011
Sunderban Wetland soil	0.05 - 12.4	Sarkar et al., 2008
Hissar Agricultural soil	2 - 51	Kumari et al., 2008
Unnao Surface soil	0.1 - 7.3	Singh et al., 2007
Thiruvallur Surface soil	75.3	Jayashree and Vasudevan, 2005
Dehradun Surface soil	326	Babu et al., 2003
Aligarh Agricultural soil	88.9	Nawab et al., 2003
Kasimedu Agricultural soil	0.1	Senthilkumar et al., 2001
Ennore Agricultural soil	2.1	Senthilkumar et al., 2001
Cochin Agricultural soil	4.8	Senthilkumar et al., 2001
Visakapatnam Agricultural soil	0.21	Senthilkumar et al., 2001
Agra Agricultural soil	0.50	Singh, 2001
Farrukhabad Agricultural soil	158	Agnihotri et al., 1996
Haridwar	61.12	Dua et al., 1996

*ND - Nondetectable level

In the sampling region, other detected classes of OCIs were aldrin, dieldrin, and endrin. The ranges of aldrin were observed from BDL - $15.28 \pm 2.6 \text{ ng g}^{-1}$. Though, these potential carcinogens present in the soil were converted into more stable compound, dieldrin (Akhil and Sujatha 2014). In the present study, the presence of dieldrin were detected at station K I ($7.71 \pm 1.1 \text{ ng g}^{-1}$) during wet season. However, traces of endrin were found in higher amounts at the station C II ($37.28 \pm 2.1 \text{ ng g}^{-1}$). The observations imply that a wide range of application of these OCIs due to their cost effectiveness and popularity (Akhil and Sujatha 2014). Long term chemical exposure in the environs has led to severe damage, consequently discovered chronic toxic effects on humans, animals, plants and aquatic organisms. Therefore, a strict ban in their production, sale and application/usage in several countries were implemented, somehow trace level residues are detected frequently in all the phases of the ecosystem owing to their persistent nature or frequent illegal usage.

4. CONCLUSION

The present research attempt focus on the monitoring assessment of OCIs present in selected soils of Palakkad district, Kerala. Among the analyzed OCIs, α -BHC, β -BHC, γ -BHC, heptachlor, aldrin, heptachlor epoxide (B), dieldrin, endrin were detected in trace amounts could be arise from recent input, however, DDT derivatives and endosulphan were found below detectable level. The levels of OCIs in the study region were found higher in the dry season than the wet season. However, gradual decrease was observed in the concentration of OCI in this region which designates a positive indication

of changing the mode practice of the synthetic chemicals towards traditional organic farming methods. Even though, the occurrence of trace amounts of OCI residues in these area signifies the importance of adopting the conventional and innovated remedial strategies for eliminating these non-biodegradable contaminants from the environment.

ACKNOWLEDGEMENT

The authors sincerely express gratitude towards the Kerala State Council for Science, Technology & Environment, Government of Kerala (Order No. 404/2013/KSCTE), for granting fund for the present work and to the Head, Department of Chemical Oceanography, School of Marine Sciences, Cochin University of Science and Technology for providing laboratory facilities.

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