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CHEMICAL ANALYSIS OF WATER SAMPLES FROM KUTTANAD WETLAND ECOSYSTEM FOR PERSISTENCE OF PYRETHROID PESTICIDE RESIDUES: LAMBDA CYHALOTHRIN, FENVALERATE AND CYPERMETHRIN

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AUTHORS' CONTRIBUTIONS

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

Kuttanad wetland is popularly considered as the "rice bowl of Kerala" where the history of paddy cultivation can be traced back to centuries. A preliminary survey revealed that various types of pyrethroid pesticides are extensively applied in Kuttanad. Lambda cyhalothrin, Fenvalerate and Cypermethrin are some of them. 5 sampling stations were selected from the rural regions of kuttanad .Two crop periods (Virippu and Puncha periods of agricultural activities) and Monsoon season were selected for the study. The water samples were collected between 7 am and 10 am The selected samples were analysed using Gas Chromatography Mass Spectrometry (GCMS). The Monsoon season and the comparatively less active cropping Virippu period followed, showed absence of residues in the surface water except for Cypermethrin. These results are suggestive of insufficiency in natural processes that degrade the pesticides released into the system due to the enormous dumping of different types of chemicals.

Keywords: Cypermethrin; kuttanad; lambda cyhalothrin; fenvalerate.

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1. INTRODUCTION

Agriculture has played a very crucial role in providing man with his basic needs such as food, fibres and other raw materials. The development of agriculture demands use of pesticides as powerful tool in protecting crops. India is the second largest pesticides producer in Asia and the twelfth largest producer globally. Since the Green Revolution, various types of toxic chemicals are being encouraged and used extensively in India. The surplus amount of these chemicals produce unwanted and unwarranted residues which pose great threat to aquatic organisms [1]. Kuttanad wetland is popularly considered as the "rice bowl of Kerala" where the history of paddy cultivation can be traced back to centuries. Most of the paddy fields in Kuttanad were reclaimed from the shallow part of the Vembanad Lake or from the periphery of the river Pamba. The fertility of the soils of Kuttanad is found to be low to medium [2]. The Monsoon floods used to enrich the soil by the deposition of the slit. The soil of Kuttanad is silty clay and is acidic in nature [3]. The main problems faced by Kuttanad rice fields are flood. lack of drainage system, intrusion of saline water and soil acidity. The developmental projects implemented as a solution to the above issues in Kuttanad vis-a-vis the Thottappally spillway, Thanneermukkom salt water barge and improvements in 'padasekharam' bunds [4-7]. These had wide range effects and impacts on the wetland ecosystem. Prevention of salt water intrusion by the Thanneermukkom regulator affected the discipline during rice cultivation season [8]. Emergence of unfamiliar diseases and pests had compelled farmers to depend more on the chemical options of pest control than the organic procedures. Non-judicious fertilizer and pesticide application has immensely polluted the ecosystem.

The pesticides applied in Kuttanad usually belong to four major categories including organochlorine, organophosphates, carbamates and synthetic pyrethroids. It was found that organophosphates and synthetic pyrethroids are widely used in the paddy fields of Kuttanad [9-12]. The advantages of these pesticides are its cost effectiveness and efficacy. The low cost often encourages farmers to go for dozes higher than recommended, thereby, affecting the environment as well as the farmers' health. A preliminary survey revealed that various types of pyrethroids are extensively applied in Kuttanad. The fact is that banned pesticides are within the reach of farmers for pests control [13-17].

Synthetic pyrethroids include a variety of insecticides which are highly toxic to fish. Lambda cyhalothrin, Fenvalerate and Cypermethrin are extensively used in the paddy fields of Kuttanad wetland. Lambda cyhalothrin is a mixture of isomers of cyhalothrin ($C_{23}H_{19}$ CIF_3 NO_3) which is degradable in alkaline water with an approximate half-life of seven days (3 – (2 – chloro – 3, 3, 3- trifluoro – 1 – propenyl) – 2, 2 – dimethyl – cyano (3 – phenoxy phenyl) methyl cyclopropane carboxylate). Fenvalerate is (RS) – alpha- cyano – 3 - phenoxy benzyl (RS) -2 – (4 chlorophenyl) – 3 methyl butyrate (C_{25} H_{22} $ClNO_3$). It is a mixture of four optical isomers which have different insecticidal activities. The 2 – S alpha (or SS) configuration known as enfenvalerate is the most active isomer with insecticidal activity. Fenvalerate consist of 23 % of this isomer. Cypermethrin is cyano – (3 – phenoxyphenyl) methyl] 3 – (2, 2 – dichloroethenyl) – 2, 2 – dimethyl cyclopropane – 1 - carboxylate (C_{22} $H_{19}Cl_2NO_3$).

The present study of Lambda cyhalothrin, Cypermethrin and Fenvalerate from synthetic Pyrethroid group were selected to determine the impact of these insecticides on the environment and the organisms which has not received much attention. The present investigation is an attempt to study the persistence of Pyrethroid pesticide residues in the aquatic ecosystem of the Kuttanad wetland.

2. MATERIALS AND METHODS

Water samples from tributary of River Pamba flowing through major areas such as Padasekharams, Valiya Pattathanam, Cheriya Pattathanam(Champakulam), Kizhakkae Kiliyan Veli, Padijarae Kiliyan Veli, Changankary and Chennamangalam (rural regions of Kuttanad) were selected; 5 sampling stations of about 1 to 1.5 km apart were chosen Fig. 1. Two cropping periods-Virippu and Puncha periods of agricultural activities and Monsoon season (off season) were selected for the study. The months selected are January, February and March during Puncha crop season; June, July and August during Monsoon and September, October and November during Virippu crop season of year 2011 . The water samples were collected between 7 am and 10 am. A quantity of 400 ml water was collected from each station and mixed together in a two litre plastic bottle; immediately taken to the laboratory for further analysis. The samples were analysed using Gas Chromatography- Mass spectrometry (GCMS) (Agilent 6890 GC/5975 MSD) following standard procedures of USEPA(1995) with the help of the ISO certified Inter- field laboratories, Cochin.

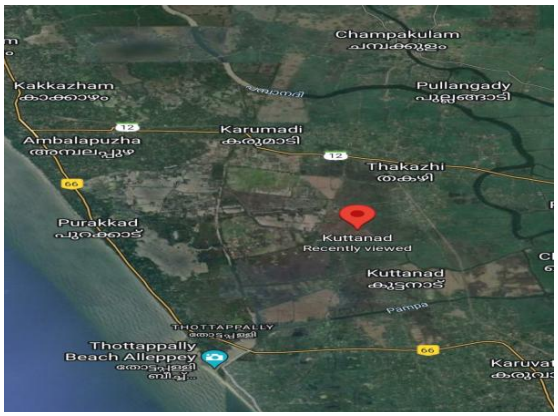

Fig .1. Map showing Kuttanad regions

3. RESULTS AND DISCUSSION

Data relating to pesticide residues in water sample are presented in Table 1 and Figs. 2 to 4.The water samples collected from the tributary of river Pampa during the various months of Puncha crop revealed the presence Lambda cyhalothrin, Fenvalerate and Cypermethrin (synthetic Pyrethroid insecticides) residues in varying concentrations. Lambda cyhalothrin and Fenvalerate residues were found to be below detectable level in the month of January but detected during February and March (Lambda cyhalothrin 21.1ppb to 19.1ppb and Fenvalerate 23.5 ppb to 20.3 ppb). (Table 1 and Figs. 2 and 3). Cypermethrin residues were detected during February (9.1 ppb) and June (8 ppb) only (Table 1 and Fig. 4). Lambda cyhalothrin and Fenvalerate insecticide residues were not detected during Monsoon and Virippu cropping (Table 1 and Figs. 1 and 2).

Persistence of insecticide residues in the tributary of river Pamba flowing through the rural regions of in Kuttanad.

Table 1. Concentration of pesticides residues in River Pamba, Kuttanad regions, 2011

Insecticides tested	Residues in ppb								
	Puncha crop			Monsoon			Virippu crop		
	Jan	Feb	March	June	July	Aug	Sept	Oct	Nov
Lambda Cyhalothrin	0	21.1	19.1	0	0	0	0	0	0
Fenvalerate	0	23.5	20.3	0	0	0	0	0	0
Cypermethrin	0	9.1	0	8	0		0	0	0

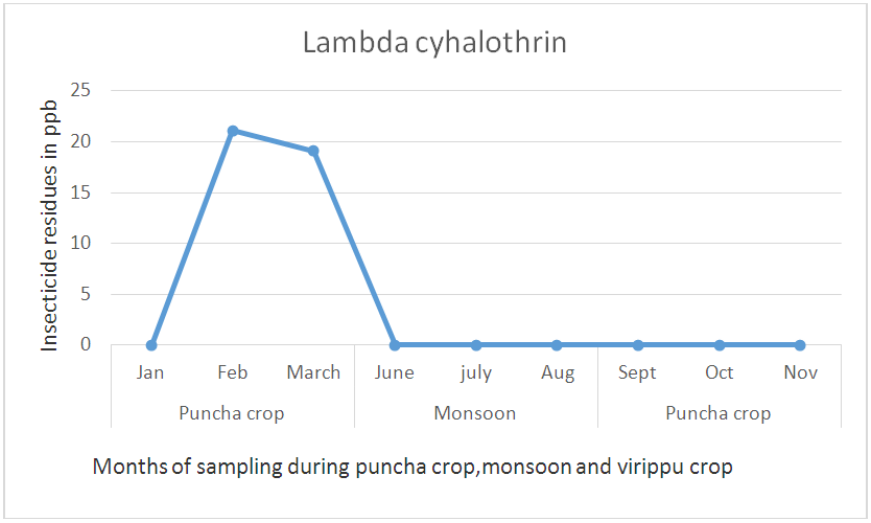

Fig. 2. Concentration of Lambda cyhalothrin residues in Kuttanad, Puncha cropping season

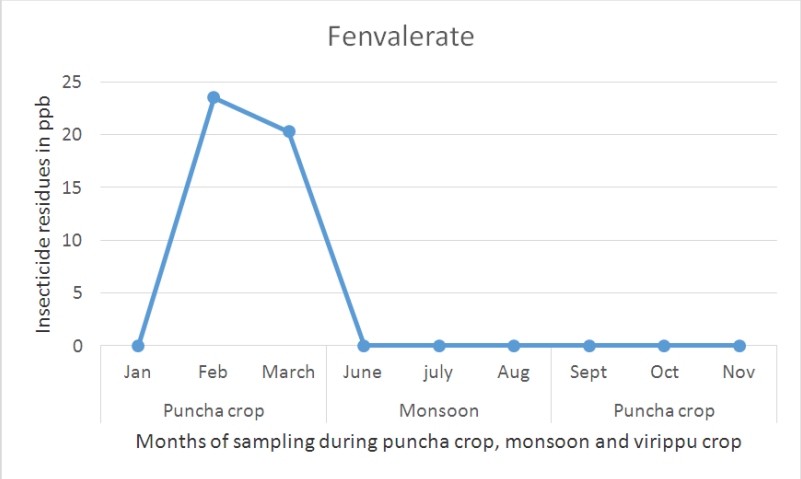

Fig. 3. Concentration of Fenvalerate residues in Kuttanad, Puncha cropping season

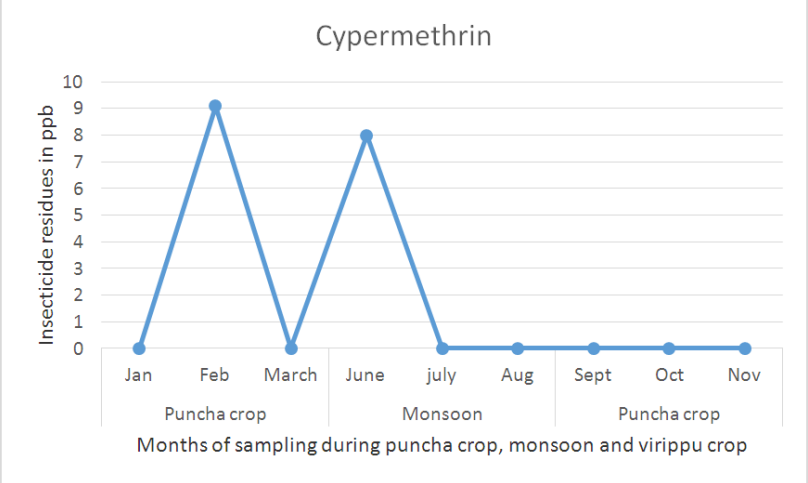

Fig. 4. Concentration of *Cypermethrin residues* in Kuttanad, Puncha cropping, *monsoon season*

The major way by which pesticides enter water body is through runoff from agricultural fields and grazing lands. Effluents from pesticide manufacturing industries stand second in contaminating aquatic environment [18]. The concentration of pesticide in surface water is closely related to the timing of its application. For example, it was determined that the maximum load of pesticide residue in surface water increase during the crop growing seasons [19,20]. The present study is in tandem with the above observation since it was found that the elevation of pesticide concentration occurred in surface water during the Puncha crop growing seasons of Kuttanad. The Monsoon season and the comparatively less active cropping Virippu period follows; show absence of residues in the surface water except for Cypermethrin.

Pesticide loss in agricultural runoff have been quantified to evaluate the contamination potential. It was estimated that they are less than 1.5 % of the pesticide applied and sometimes less than 0.1 % [19,21]. Comparison with the original concentration of the applications estimates may be less, but their concentrations in water can reach levels injurious to the various organisms inhabiting the water medium. Concentration of pesticides in surface water was highest in late Spring and early Summer, following the Spring application [22]. Another entry route of pesticides into water body is through atmospheric long range transport with the help of wind [23,24]. It was found that spray droplets smaller than10 µm can be transported over many kilometres by air at low wind speed during application and their movement is favoured by a continuous decrease in droplet size due to evaporation [25,26] When an insecticide is introduced as a spray near a water body, the main route to surface water is spray drift. Spray drift is a complex problem in which the design of the

equipment used, its condition, application parameters, spray, physical properties and formulation and metrological conditions greatly influence the pesticide loss [27].

In the present study, late summer and early pre Monsoon seasons showed the presence of insecticide residues of Lambda cyhalothrin, Fenvalerate and Cypermethrin. Cypermethrin was the only insecticide detected during Monsoon season. The selected insecticide residues were absent during the Virippu cultivation. Puncha cultivation period was a period of hectic agricultural activities but the Virippu cultivation was less enthusiastic during the period of study. Most of the farmers in the area were less interested in cultivation during Virippu cropping season due to many socio-economic reasons. Thus Mboth monsoon season and Virippu cropping period were comparatively free of paddy cultivation and insecticide application. Lambda cyhalothrin and Fenvalerate are extensively used in various trade names in the Padasekharams of Kuttanad wetland.

A survey conducted in the present study has revealed an element of neglect and ignorance regarding insecticide application on the part of farmers and labourers in the region. Poor operator handling is a major reason for surface water entry of pesticides [28,29]. Repairing the sprayer while spraying, neglecting instructions and using faulty or wrong instruments can increase the chances for direct water contamination with higher doses of pesticides [30,31]. The amount of pesticides in surface water as observed in the present study may also be due to the above mentioned procedural errors.

Lambda cyhalothrin, Fenvalerate and Cypermethrin were found in varying concentrations during the period of hectic agricultural activities. The presence of Pyrethroid insecticide residues during February and March revealed its widespread application. However, investigations after two months regarding their persistence were not detectable in the aquatic medium. The possible dilution of residues during Monsoon shows absence of paddy cultivation during Monsoon season, comparative less intense farming during Virippu cropping and various pesticide degradation mechanisms, are the probable reasons for the absence of these insecticide residues in the surface water. The presence of Cypermethrin in water during the Monsoon season can be correlated to its direct application to water for mosquito larvae control.

Several studies have been conducted to reveal the presence and quantity of insecticides in surface water. An exhaustive study of pesticides in water was initiated in 1968 by the United States Geological Survey Agency in October 1968 to September 1971. The development of synthetic Pyrethroids as broad spectrum commercial insecticide around 1980 represented a new threat to fishes. Naturally occurring Pyrethrins from a Eurasian species of Chrysanthemum used for decades were found safe and none consistent. They seldom have enough persistence to reach water from a normal application. Chemical substitution of 3 or 4 labile group in the basic Pyrethrin molecule by scientists paved way for invention of more stable and more potent synthetic analogs of natural Pyrethrins [32-35]. The introduction of more stable side-chains and rings, as well as the inclusion of bromine, chlorine and fluorine resulted in synthetic pyrethroids that are less susceptible to oxidation, hydrolysis and photolytic reactions. Synthetic Pyrethroid insecticides have innate toxicity to fishes and have longer persistence, severe lipophilicity and their LC50 values were similarly low for numerous chlorinated hydrocarbons against a variety of fish species and life stages [36].

There has been a dramatic increase in the use of Pyrethroid pesticides to control insect pests in recent years [37]. According to Anwar et al. [38] Pyrethroid insecticides are highly toxic, thus, their chances to accumulate in the body of aquatic organisms is great. They are extremely hydrophobic and highly lipophilic in nature [39] and their degradation products are more toxic than their parent compounds. Boxall et al. [40] Laskowski et al. [41] indicates that Lambda cyhalothrin, a Pyrethroid pesticide has a high potential of bio concentrates. The gills of fish easily absorb pyrethroids because of the lipophilic nature of the pesticide [42]. The acute toxicity of Pyrethroids to fish is usually measured in filtered or purified water. Such studies have demonstrated Pyrethroids as toxic as organochlorine insecticides. But later studies in the field suggests that they are less potent than expected from laboratory studies. It was determined that Pyrethroids have extremely low water solubility and high affinity for particulate matters in solution, thus, not readily vailable for uptake by fishes in the field pond [29,41].

Pyrethroid pesticides are the derivatives of chrysanthemic acid, containing an isobutenyl group, and are very much susceptible to photochemical oxidation. They undergo photolysis by hydrolysis of the ester bond, cis-trans isomerization, carboxylation, and reductive dehalogenation [3]. California drainage systems are of special concern because of the high use of Pyrethroids and persistence of the residue in sediment [43]. Most of the channels in Kuttanad region are almost covered with various types of weeds and thus, sunlight is hindered from reaching the

surface water. This will negatively affect any kind of photolytic degradation of pesticides. According to Rathore et al. [27], microbial degradation of Pyrethroid pesticides is a very slow process. This study shows persistence of Lambda cyhalothrin and Fenvalerate in the Puncha crop; Cypermethrin was found during both Puncha cultivation and Monsoon season. These results are suggestive of insufficiency in natural processes to degrade the pesticides released into the system due to the enormous dumping of different types of chemicals.

The Kuttanad wetland system has own its unique natural features which discouraged the spreading of weeds and the explosion of pest populations. However, drastic human interferences into the ecosystem which began with the construction of the Thanneermukkom bund altogether transformed almost all self-regulative healing mechanisms of the wetland. The existence of this structure has altered majority of the hydrographic parameters in the ecosystem. Undiscerning introduction of different types of insecticides and their unscrupulous application in the field to attain more yield has resulted in insecticide resistant insect population explosion, appearance of strange pests and unfamiliar diseases. Insecticides will also adversely affect the natural enemy population of the different pests in the ecosystem. Cypermethrin and Lambda cyhalothrin have significantly reduce spiders population in the rice fields [44,25].

There are reports from various parts of the world regarding the presence and chances of bio-accumulation, bio-concentration and bio-magnification of pesticides when pesticide residues are available in water. Most of the studies carried out concentrated on the organochlorine pesticides. Nayak et al. [3], Protasowicki et al. [18], Zhou R et al. **[**20], Manahan **[**44], Biego et al. [46], Mahboob et al. [46] found out that pesticide residues are present in the muscles of Cirrhinus mrigala collected from farms and River Chenab at Trimus Head and Jhang. Endosulfan, Carbofuran, Diazinon, Parathion methyl, Dimethoate, Malathion, Chlorpyrifos, Cypermethrin, Carbosulfan, Methamidophos and Isoproturon were detected in their study.

4. CONCLUSION

The presence of pesticides in the surface water as revealed in the present study is an indication that insecticide residues get incorporated in the body of aquatic organisms inhabiting the area. The inhabitants in Kuttanad have an in-depth relation with the water around them. The study conducted among the people of Kinakary and Kuttanad has convincingly proven that 20 % of the people that participated in the study use water taken directly from the lake and rivers for drinking and cooking purposes. Nearly 100 % are using this water for washing, bathing and cleaning. Hence, slightest amount of insecticides in the surface water is of great concern. Insecticide residues present in the aquatic and amphibious organisms must be taken serious because they constitute major share in the diet of the people living in and around Kuttanad. Ducks reared in this region have emerged as a brand (Kuttanaden Tharavu) and lavishly served in form of various recipes in resorts and star hotels. The hotels and resorts in this area depend mostly on the water collected from the wells, ponds and rivers located nearby. A number of roadside vendors are also involved in selling live ducks and freshly collected fishes from the field. Ducks and fishes are cleaned and dressed on demand r from the paddy fields' water situated along the wayside. In these circumstances the possibility of pesticide entry into human diet is a matter of serious concern. Thus the chances of insecticide residue entry at different trophic levels and their consequences call for elaborate investigations.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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