



Assessment of Organochlorine (OC) and Organophosphorus (OP) pesticide residues in Groundwater near Agricultural activity areas of East Godavari District, Andhra Pradesh State, India

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ABSTRACT:

In this research study, thirty-eight (38) organochlorine (OC) and twenty (20) organophosphorus (OP) pesticides in groundwater samples of 3 research study areas near agricultural areas of konaseema region, East Godavari district, Andhra Pradesh state, India were investigated as per acceptable analytical methods. Our research findings confirmed the presence of the 38 OCP's and 20 OPP's of different concentrations in groundwater. Gas Chromatography-Mass Spectrometry (GC-MS) was employed to measure the pesticide residual concentrations in groundwater samples. The mean concentration level of the OCP's and OPP's in groundwater samples were between ND-1300 µg/l and 30-1080 µg/l respectively in research area-1. The concentration level of OCP's and OPP's in water samples were between ND-1280 µg/l and ND-970 µg/l respectively in research area-2. In research area- 3, the levels of OCP's and OPP's were between ND-1250 µg/l and ND- 1150 µg/l respectively. A higher concentration of OC pesticide residues were detected in research area -1 while higher concentrations of OPP's were detected in research area-3.

1. Introduction

Application of fertilizers and pesticides can enhance agriculture production but can cause harmful impact on water quality [1-3]. The sources for water pollution are mainly from pesticide in agricultural activities, industrial activities and domestic wastes etc. [4, 5]. Usage of Pesticides and veterinary drugs in agricultural activities result in subsurface water contamination [6-8]. The contamination of environment is due to hazardous substances such as persistent organochlorine pesticides and organic pollutants have become a worldwide public health concern [9]. Agricultural practices resulted in serious impacts on the environment, causing an increase in the level of pesticide residues in natural water, soil, river sediments and foodstuffs [10, 11]. But synthetic organochlorine pesticides have significant usage in global level and the OCP's are persistent organic pollutants which can cause environmental problems

[12-14]. These residues on reaching natural water bodies disturb flora and fauna. Pesticides which do not mortify easily or take time to mortify are more harmful [15]. HCHs isomers are found to be wide-ranging toxicants which accumulate in food chain with high risk to the environment [16]. Pesticides like DDT and HCH were used extensively in India for agricultural purposes. Pesticides used in agricultural soils will be transported by some means and reach the groundwater sources which can cause health hazards among humans and also to wild life [17- 19]. It is estimated that nearly 25000 MT of chlorinated pesticides are in usage annually in India [20]. Bad agricultural practices can lead to contamination of surface and ground waters with pesticides [21, 22]. The high degree of toxicity of pesticides may cause skin irritation, respiratory problems, dizziness and nausea. The lower toxic impacts of pesticides are associated with various chronic diseases like nervous system problems,



parkinson's disease and abnormal immune system [23]. A. Akinnusotu et al. [24] researched on OC pesticides and the pesticide concentrations were higher in soil near the river which can cause adverse effect to aquatic life by bio-accumulation. The residues of various pesticides reach the groundwater sources through the process of leaching; careless disposal of empty containers and washings of equipment etc. [25]. The over use of pesticides has brought an increased environmental concern [25 -27]. The World Health Organization consultation developed guideline values for nine herbicides in drinking water based on evaluation of each compound's toxicity data. According to WHO's calculation, an average adult of 70 kg consumes 2 l/day water and that 10% of the average daily intake is allocated to drinking water [28]. Even though the pesticides provide some benefits but they can cause hazardous consequences [29 -31]. The high detection frequencies of OCPs in soil samples indicated their wide spread in agricultural soil with DDTs, HCHs and Endosulfans being the most dominant OCPs [32]. The main objective of the present study is to characterize ground water for assessment of concentrations of pesticide residues to estimate the toxicity of ground water near agricultural areas.

2. Materials and Methods

The Research areas selected

The research areas for the present study includes Nedunuru village of Ainavilli mandal (81.9515053 N, 16.641359 E), Palagummi village of Amalapurammandal (81.9599204 N, 16.6172554 E) and K.Pedapudi village (81.9355851 N, 16.6191004 E) of Ambhajipetamandal in East Godavari district of Andhra Pradesh state, India. The study areas were located nearer to the hectic agricultural activity. The study area maps are shown in Figure-1.

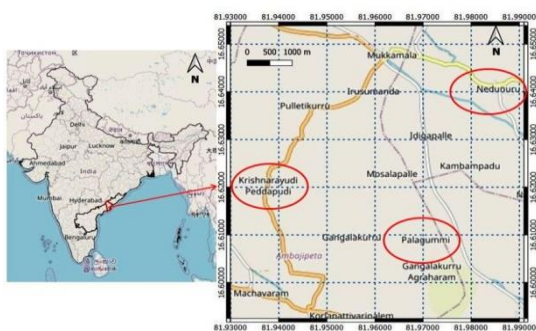


Fig. 1: The study region and sampling locations

Groundwater sampling

From each ten Groundwater samples 3 representative samples were prepared and from each village of the three study areas in pre and post monsoon seasons during the months of April-May and November-December. The samples were collected as per the standard procedures [33, 34]. To establish the quality data, strict quality assurance and quality control programme has been followed for the pesticides.

Determination of organochlorine (OC) and organophosphorous (OP) pesticides

Gas chromatography mass spectrometry (GC-MS) was employed to detect the pesticide residues in groundwater. GC CLARUS 550 PerkinElmer system comprising a gas chromatograph interfaced to a mass spectrometer (GC-MS) instrument is used with the following conditions.

Equipment used: Thermo GC – Trace Ultra Ver. 5.0
Thermo MS DSQ II

Column : DB 35 – MS capillary standard non – polar column

Dimensions : 30m, ID: 0.25mm, film: 0.25µm

Carrier gas : He, flow: 1.0 ml/min

Temperature : oven Temp 80°C to 250°C at 10°C/min

Gas chromatography conditions

Pesticide residues were separated from the sample matrix by passing inert gas helium through the matrix. The sample was placed in a sealed container and left at a constant temperature until the gas and liquid phases are in equilibrium. The target substances in the gas phase (headspace) were collected by gas tight syringe. This was injected into the GC/MS. Calibration curve were made by dissolving the target chemicals in purified water, and then treated in the same manner. The target, volatile compounds were desorbed from the aqueous phase to the gas phase (purged) and are then separated from the stream of gas (trapped) by adsorbent filters. The adsorbent material was then heated in a stream of GC carrier gas (usually pure helium). This releases the trapped substances into the carrier gas and the target analytcs were introduced to GC-MS [35].

3. Results & Discussion

The water samples collected from the nearby areas of agricultural activities in the study areas of Nedunuru, Palagummi and K. Pedapudi villages in konaseema region of East Godavari District; Andhra Pradesh were analyzed by GC-MS for assessing the levels of pesticide



residue contamination. 58 pesticides were employed for agricultural activity in 3 study areas by the farmer community. Out of 58 pesticides, 10 pesticides were employed by the farmers in Nedunuru village (Research area -1), 27 pesticides were found to be used in Palagummi village (Research area -2) and 21 pesticides were used by the farmers in K.Pedapudi village (Research area -3).

Research area- 1:

The concentration levels of organochlorine and organophosphorous pesticides of research area-1 (Nedunuru village) during pre and post monsoon seasons and their mean concentrations are presented in table -1 and in figures 2(a), 2(b) and 2(c) respectively. Results showed that the presence of 10 compounds, out of which 8 compounds were organochlorine and 2 compounds were organophosphorus.

OC pesticides: It was recorded that the mean concentrations of terbuthylazine, dimethachlor and bifenthrin were 1290 µg/l, 1300 µg/l and 1220 µg/l respectively in samples 1, 2 and 3. In all samples it crossed the guideline values of 7 µg/l, 20 µg/l and 20 µg/l [36- 38]. The average concentration of simazine and acetochlor were noted as 130µg/l, 655µg/l and 330 µg/l, 165 µg/l respectively in samples 1 and 2, which crossed the guideline value of 20 µg/l [39, 40]. In sample- 3 their concentrations were below the detectable limit. The concentration of heptachlor epoxide in samples 1 and 3 was found to be 570 µg/l and 980 µg/l respectively and in sample-2 it was not detected. The mean concentration of propachlor in samples 1 and 3 was observed as 20 µg/l, while in sample -2 it was 10 µg/l. In all samples it was below the guideline value [39]. The concentration of iprodione in samples 1 and 2 was noted as 240 µg/l where as in sample 3 it was 120 µg/l.

OP pesticides: The phosmet and ethion concentration in samples 1, 2 and 3 were found to be 90 µg/l, 45 µg/l, 30 µg/l and 650 µg/l, 980 µg/l, 1080 µg/l respectively.

Research area- 2:

The concentration levels of organochlorine and organophosphorous pesticides of Area- 2 (Palagummi village) during pre and post monsoon seasons are presented in table -2 and in figures 3(a), 3(b) and 3(c) respectively. Results showed that the presence of 27 compounds, out of which 19 compounds were

organochlorine and 8 compounds were organophosphorus.

OC pesticides: Higher concentration of bifenthrin and atrazine in sample- 4 were observed at 1260 µg/l and 640 µg/l while in samples 5 and 6 they were noted as 970µg/l and 1280 µg/l. They crossed the guideline value of 100µg/l [37]. Heptachlor epoxide, cyproconazole, dieldrin were found to be 980 µg/l, 10 µg/l and 10 µg/l in sample- 4 while in samples 5 and 6 they were below the detectable limit. The concentration of cypermethrin in sample- 4 was 120µg/l while in samples 5 and 6 it was 655 µg/l. The pesticide residue, aldrin in sampling location of 4 was observed at 504 µg/l and in samples 5 and 6 it was below the detectable limit. The concentration of deltamethrin in sample- 4 was 100µg/l while in samples 5 and 6 it was observed as 750µg/l. The concentrations of fluvalinate, chlorpyrifos methyl, P,P –DDT were detected in samples 5 and 6 and their concentrations were 40µg/l, 30µg/l and 20µg/l respectively. Fipronil and permethrin were detected only in sample- 6 of concentrations 270µg/l and 10µg/l respectively. The concentration of propanil was identified in samples 5 and 6 as 35 µg/l and 70µg/l respectively. Metalachlor and hexachlorobenzene were detected in sample- 6 only whose concentrations were 285 µg/l and 105 µg/l. Both propanil and metalachlor crossed the guideline value of 10µg/l [39]. The concentration of fenvalerate in samples 5 and 6 was observed at 645 µg/l. The concentration of procymidone in sample- 4 was 320µg/l while in the case of samples 5 and 6 it was observed at 230µg/l. The concentration of captan in all the three samples was 20µg/l.

OP pesticides: The concentration of ethion in sample-4 was found to be 970 µg/l while in samples 5 and 6 it was 320 µg/l. The mean concentrations of dimethoate and prothiofos in samples 4, 5 and 6 were 60 µg/l and 30 µg/l respectively. It crossed the guideline value of 6 µg/l [39]. The concentration of phosmet in sample- 4 was observed as 30 µg/l while in samples 5 and 6 it was noted as 5 µg/l. The concentration of methamidophos in samples 5 and 6 was 90 µg/l. Triazophos concentration in sample -4 was noticed as 20 µg/l whereas in samples 5 and 6 it was 40 µg/l. In all the three samples the concentration of azinphos ethyl was identified as 5 µg/l. The concentration of phosalone in samples 5 and 6 was observed to be 10µg/l.

**Research area- 3:**

The concentration levels of different pesticides present in groundwater near agricultural areas of area- 3 during pre and post monsoon seasons are presented in table -3 and in figures 4(a), 4(b) and 4(c) respectively. Results showed that the presence of 21 compounds, out of which 11 compounds were organochlorine and 10 compounds were organophosphorus.

OC pesticides: The concentration of atrazine in samples 6 and 7 was observed at 870 μ g/l. It crossed the guideline value of 100 μ g/l [36, 38]. The concentration of alachlor was found in sample-6 as 40 μ g/l. It crossed the guideline value of 20 μ g/l [36]. In samples 8 and 9, the concentration of butachlor was observed at 1250 μ g/l and 1280 μ g/l respectively. It crossed the guideline value [38]. The mean concentration of dichlorvos in all samples was 30 μ g/l. The concentration of lambda cyhalothrin in samples 7 and 8 was observed as 120 μ g/l. The aldrin concentration in samples 8 and 9 was 920 μ g/l. The concentration of δ -HCH in samples 7, 8 and 9 was observed as 620 μ g/l, 50 μ g/l and 20 μ g/l respectively. The concentrations of β -HCH, lindane, alachlor and heptachlor were found in only one sample as 1180 μ g/l, 1250 μ g/l, 40 μ g/l and 10 μ g/l respectively. The concentration of dieldrin in sample-9 was observed as 920 μ g/l. The higher concentration of the residues of HCH isomers show their leaching behavior [41, 42]. The water samples collected from wetland system, Kerala, showed higher concentration of HCH isomers [43, 44].

OP pesticides: The concentration of chlorpyrifos in samples 6, 7 and 8 was 340 μ g/l, 60 μ g/l and 30 μ g/l respectively. In all the three samples, phorate and fenitrothion concentrations were observed at 20 μ g/l each. Phosphomidon concentration in sample 8 was observed at 720 μ g/l. The concentration of parathion in samples 8 and 9 was 20 μ g/l and 30 μ g/l respectively. The concentrations of profenophos and quinalphos in sample-9 were observed at 120 μ g/l and 860 μ g/l respectively. In all the samples it crossed the guideline value [36, 38]. Diazinon, methyl parathion were found in only one sample as 1150 μ g/l and 90 μ g/l respectively. The mean concentration of monocrotophos in all samples was 40 μ g/l.

Conclusions

Based on the detected concentrations of 38 OCP's and 20 OPP's in ground water in the study areas, it can be concluded that the ground water near agricultural

areas are contaminated with Organochlorine and Organophosphorous pesticides. The research results revealed that the ground water is significantly contaminated by OC pesticides compared to OP pesticides.

Conflicts of interest

The authors declare that they have no conflicts of interest.

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**Table 1.** Organochloro and organophosphorous pesticide residues in Area-1(Nedunuru village).

S.No	Name of the Pesticide	Concentration (µg/l)										
		Sample -1			Mean	Sample- 2			Mean	Sample- 3		
		Monsoon		Monsoon		Monsoon						
		Pre	Post	Pre		Post	Pre	Post		Pre	Post	Mean
Organochlorine pesticides												
1	Dimethachlor	1300	1300	1300	1300	1300	1300	1300	1300	1300		
2	Simazine	130	130	130	1290	20	655	ND	ND	ND		
3	Heptachlor Epoxide	830	310	570	ND	ND	ND	980	980	980		
4	Terbuthylazine	1290	1290	1290	1290	1290	1300	1290	1290	1290		
5	Acetochlor	330	330	330	330	ND	165	ND	ND	ND		
6	Propachlor	20	20	20	20	ND	10	20	20	20		
7	Bifenithrin	1220	1220	1220	1220	1220	1220	1220	1220	1220		
8	Iprodione	240	240	240	240	240	240	ND	240	120		
Organophosphorous pesticides												
9	Phosmet	90	90	90	90	ND	45	60	ND	30		
10	Ethion	980	310	645	980	980	980	1080	1080	1080		

ND: Not detected

Table 2. Organochloro and organophosphorous pesticide residues in Area -2 (Palagummi village).

S.No	Name of the Pesticide	Concentration (µg/l)										
		Sample- 4			Mean	Sample- 5			Mean	Sample- 6		
		Monsoon		Monsoon		Monsoon						
		Pre	Post	Pre		Post	Pre	Post		Pre	Post	Mean
Organochlorine pesticides												
1	Bifenithrin	1260	1260	1260	970	970	970	970	970	970		
2	Atrazine	ND	1280	640	1280	1280	1280	1280	1280	1280		
3	Cypermethrin	240	ND	120	1270	40	655	1270	40	655		
4	Aldrin	1080	ND	540	ND	ND	ND	ND	ND	ND		
5	Heptachlor Epoxide	980	980	980	ND	ND	ND	ND	ND	ND		
6	Procymidone	320	320	320	230	230	230	230	230	230		
7	Deltamethrin	20	ND	10	750	750	750	750	750	750		
8	Captan	20	20	20	20	20	20	20	20	20		
9	Cyproconazole	30	ND	10	ND	ND	ND	ND	ND	ND		
10	Dieldrin	20	ND	10	ND	ND	ND	ND	ND	ND		
11	Fluvalinate	ND	ND	ND	40	40	40	40	40	40		



12	Chloropyrifos methyl	ND	ND	ND	30	30	30	30	30	30
13	P,P - DDT	ND	ND	ND	20	20	20	20	20	20
14	Hexachloro benzene	ND	ND	ND	ND	ND	ND	ND	ND	ND
15	Fenvalerate	ND	ND	ND	ND	1290	645	ND	1290	645
16	Fipronil	ND	ND	ND	ND	ND	ND	540	ND	270
17	Permethrin	ND	ND	ND	ND	ND	ND	20	ND	10
18	Metolachlor	ND	ND	ND	ND	ND	ND	ND	570	285
19	Propanil	ND	ND	ND	70	ND	35	70	70	70
Organophosphorous pesticides										
20	Ethion	970	970	970	320	320	320	320	320	320
21	Dimethoate	60	60	60	60	60	60	60	60	60
22	Phosmet	60	ND	30	10	ND	5	10	ND	5
23	Prothiofos	30	30	30	30	30	30	30	30	30
24	Triazophos	20	20	20	40	40	40	40	40	40
25	Methamidophos	ND	ND	ND	ND	180	90	ND	180	90
26	Azinphos ethyl	ND	10	5	10	ND	5	10	ND	5
27	Phosalone	ND	ND	ND	10	10	10	10	10	10

ND: Not detected

Table 3: Organochloro and Organophosphorous pesticide residues in Area -3 (K. Pedapudi village)

S.No	Name of the Pesticide	Concentration ($\mu\text{g/l}$)										
		Sample- 7			Mean	Sample- 8			Mean	Sample- 9		Mean
		Monsoon		Monsoon		Monsoon						
		Pre	Post	Pre	Post	Pre	Post	Pre	Post			
Organochlorine pesticides												
1	Lindane	1250	1250	1250	ND	ND	ND	ND	ND	ND		
2	Butachlor	ND	ND	ND	1250	1250	1250	1280	1280	1280		
3	β -HCH	1180	1180	1180	ND	ND	ND	ND	ND	ND		
4	Atrazine	870	870	870	870	870	870	ND	ND	ND		
5	Aldrin	ND	ND	ND	920	920	920	920	920	920		
6	δ -HCH	620	620	620	50	50	50	20	20	20		
7	Lambda cyhalothrin	120	120	120	120	120	120	ND	ND	ND		
8	Alachlor	80	ND	40	ND	ND	ND	ND	ND	ND		
9	Dieldrin	ND	ND	ND	ND	ND	ND	920	920	920		
10	Dichlorvos	30	30	30	30	30	30	30	30	30		
11	Heptachlor	20	ND	10	ND	ND	ND	ND	ND	ND		



Organophosphorous pesticides										
12	Diazinon	1150	1150	1150	ND	ND	ND	ND	ND	ND
13	Chlorpyrifos	340	340	340	60	60	60	30	30	30
14	Phosphomidon	ND	ND	ND	720	720	720	ND	ND	ND
15	Methyl parathion	90	90	90	ND	ND	ND	ND	ND	ND
16	Monocrotophos	40	40	40	40	40	40	40	40	40
17	Phorate	20	20	20	20	20	20	20	20	20
18	Fenitrothion	20	20	20	20	20	20	20	20	20
19	Quinalphos	ND	ND	ND	ND	ND	ND	860	860	860
20	Profenophos	ND	ND	ND	ND	ND	ND	120	120	120
21	Parathion	ND	ND	ND	20	20	20	30	30	30

ND: Not detected

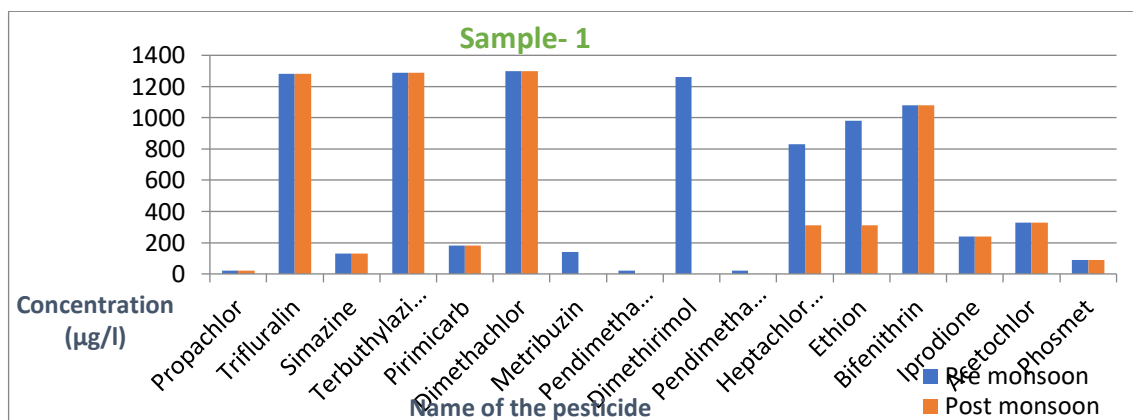


Figure- 2(a)

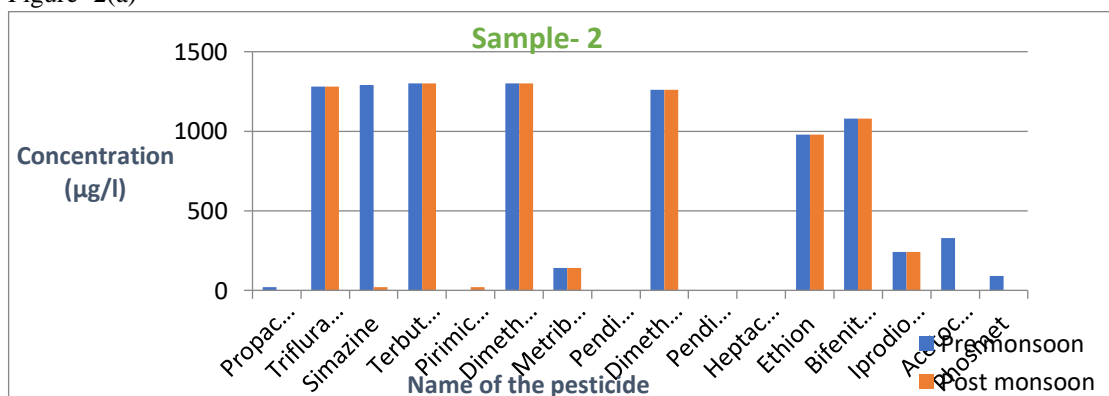


Figure- 2(b)

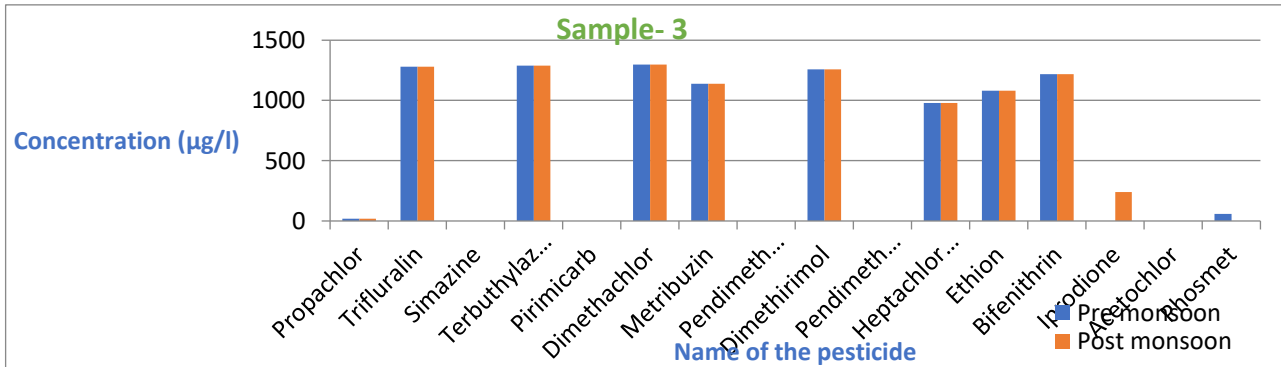


Figure- 2(c)

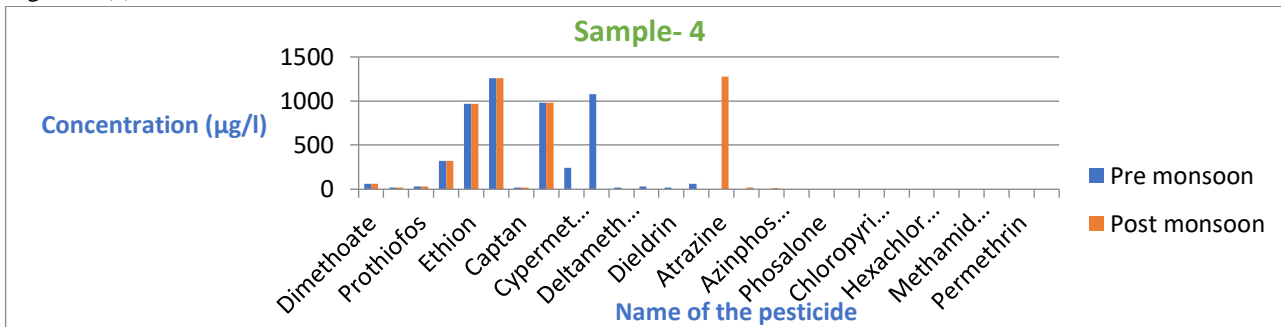


Figure- 3(a)

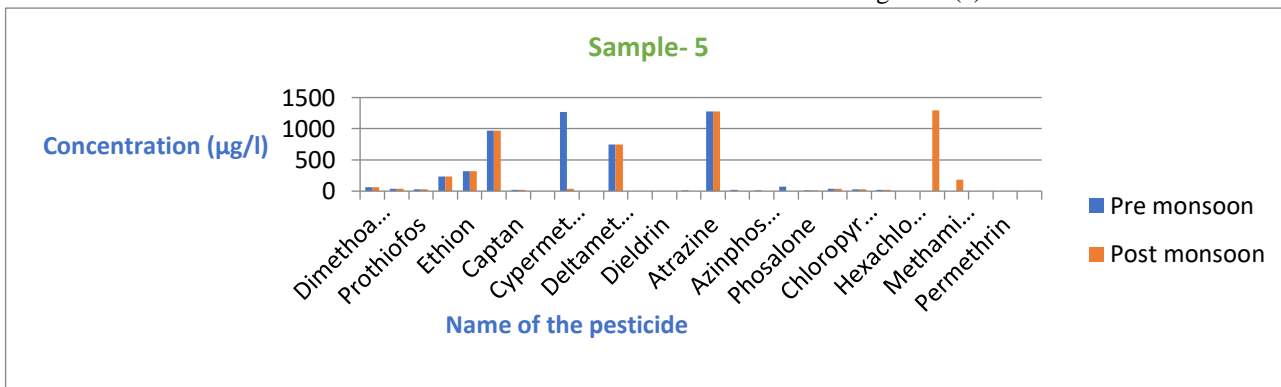


Figure- 3(b)

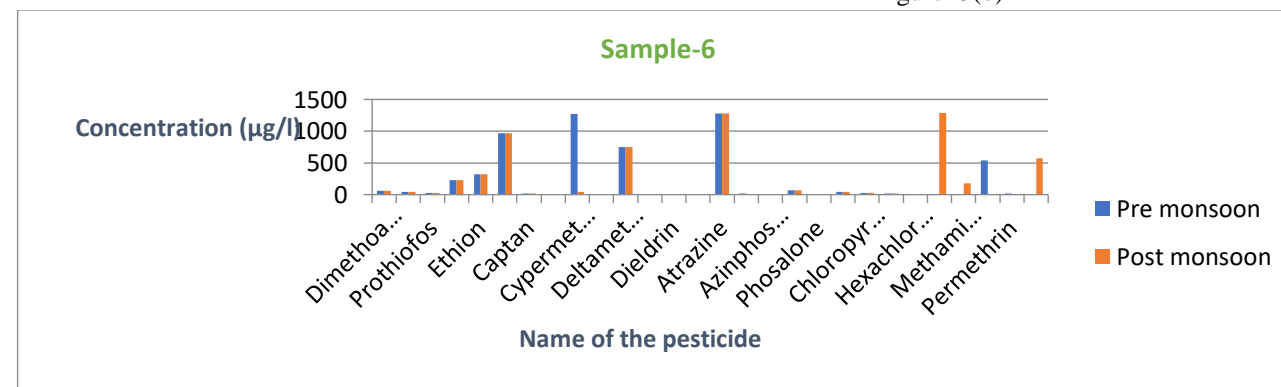


Figure- 3(c)

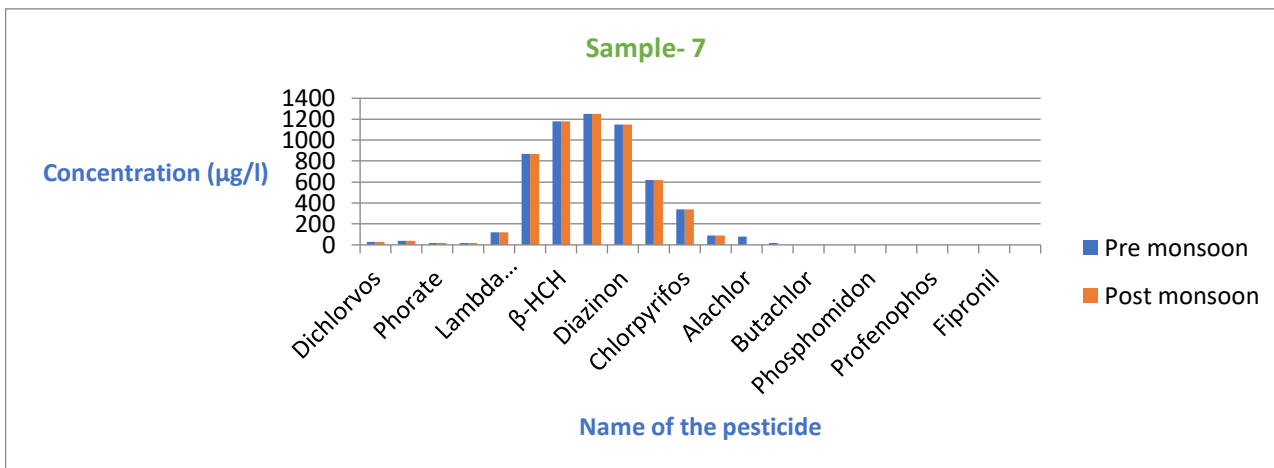


Figure- 4(a)

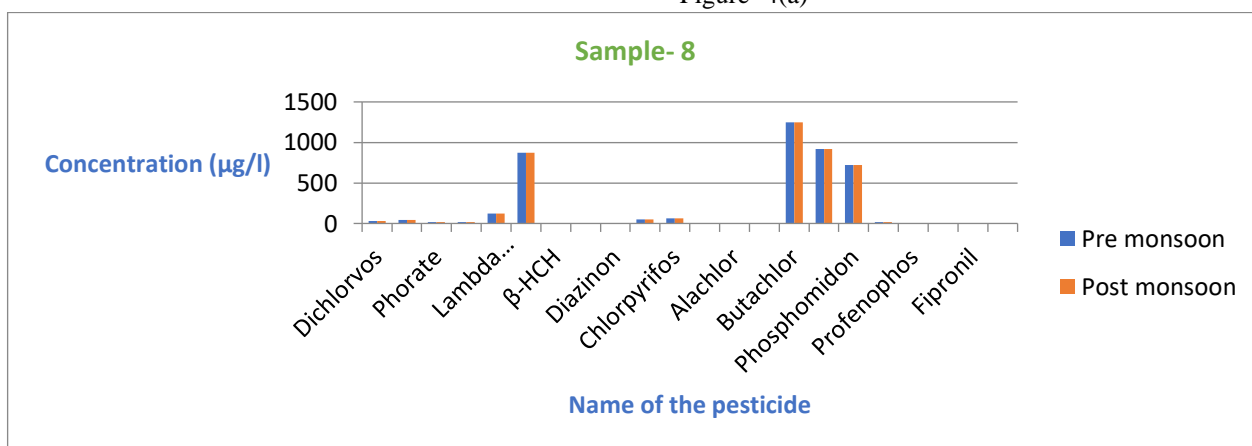


Figure- 4(b)

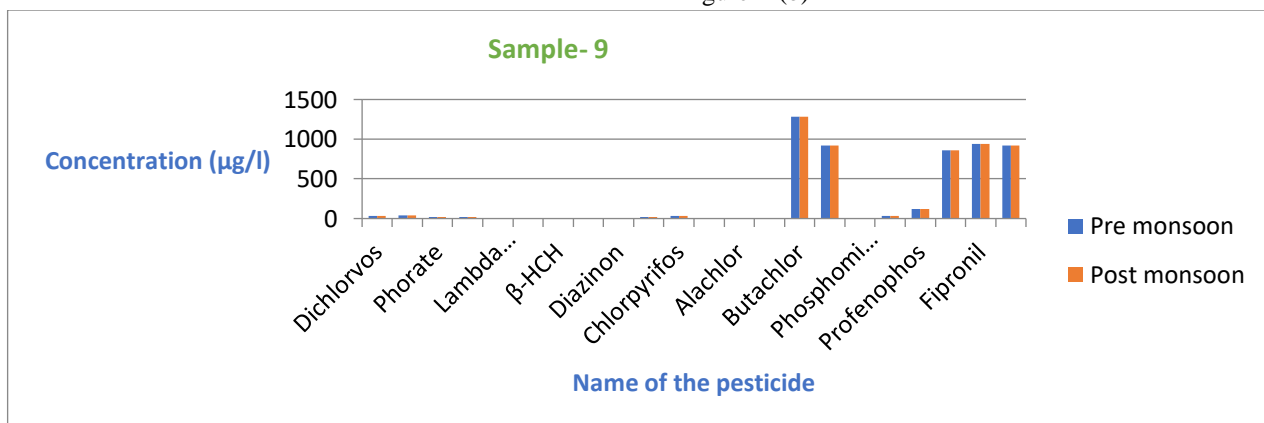


Figure- 4(c)