International Journal of Research in Chemistry and Environment
Vol. 1 Issue 1 July 2011(77-84)
ISSN 2248-9649

Research Paper

Organochlorine and Organophosphorous pesticides residues in Water of River Ganga at Bhagalpur, Bihar, India

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Available online at: www.ijrce.org

(Received 18th May 2011, Accepted 27th May 2011)

Abstract *- River Ganga along with its tributaries is the largest and also a very important river basin of India. The river Ganga, a holy river has over the years been subjected to tremendous pressure due to discharge of untreated sewage, industrial effluents, residues of pesticides and insecticides used in farms which has adversely affected the ecological health of the river and its basin The objective of this study was to identify and quantify some selected pesticides both organochlorine and organophosphorous present in water of River Ganga at Bhagalpur. These pesticides are mainly used to control the soil and crop pests in agricultural fields and direct dumping of wastes in to river systems. Combination of their physico-chemical properties such as low aqueous solubility, moderate vapor pressure and octanol – water partition coefficient and persistence in the environment make them capable of long-range transport. Water samples from 3 sites (up stream, mid stream and down stream) were collected and analyzed for their pesticide profile. The study has shown the presence of both organochlorine and organophosphorous pesticides in the water of River Ganga at Bhagalpur. Liquid-liquid extractions followed by GC-ECD were used for the determination of these compounds. Among the various pesticides analyzed, high concentration of Methyl parathion, Endosulfan and DDT were observed in water samples collected from the River Ganga at Bhagalpur. Especially, the concentration of DDT was quite high which might be due to slow degradation of DDT in soil i.e. 75 –100% in 4-30 years. The present study was first attempt to identify and quantify some selected pesticides in water and sediment of the river Ganga at Bhagalpur, and may be important as the Bhagalpur city gets its water supply from the river for drinking purposes.*

Key words: Ganga; Bhagalpur; Organochlorine; Organophosphorous; Pesticides; Water; Sediment etc.

Introduction

Several hundred pesticides of different chemical nature are currently used for agricultural purposes all over the world. Because of their widespread use, they are detected in various environmental matrices, such as soil, water and air. Pesticides are divided in to many classes, of which the most important are organochlorine and organophosphorous compounds. Organochlorine pesticides are known to resist biodegradation and therefore they can be concentrated through food chains and produce a significant magnification of the original concentration at the end of the chain. It has been cited that the degradation of DDT in soil is 75-100% in 4-30 years. Due to long residence time of these substances in the environment, there is a great interest in examining the pollution they cause. Oraganophosphorous pesticides, on the other hand, are known to degrade rapidly depending on their formulation method of application, climate and the growing stage of the plant.

The general progression of pesticide development has moved from highly toxic, persistent and bio-accumulating pesticides such as DDT, to pesticides that degrade rapidly in the environment and are less toxic to non-target organisms. The developed countries have banned many of the older pesticides due to potential toxic effect to man and\or their impact on ecosystems, in favour of more pesticide formulations. In the developing countries, however, organochlorine pesticides still remain the cheapest to produce and, for some purposes, remain highly effective. Developing countries maintain that they cannot afford, for reason of cost or efficacy, to ban certain older pesticides. The dilemma of cost/efficacy, versus ecological impacts, including long range transport, and access to modern pesticides formulations at low cost remains a contentious global issue. Pesticides like DDT and HCH were used extensively in India till recently both for agricultural and sanitary purposes. It is estimated that about 25000 MT of chlorinated pesticides were used annually in India and DDT accounted for 40% of this group [1]. Pesticide residues reach the aquatic environment through direct runoff, leaching, and careless disposal of empty container, equipment washing etc [2]. Surface water contamination may have ecotoxicological effects for aquatic flora and fauna as well as for human health if used for public consumption [3, 4, 5, 6] .In India, the concentration of these pesticides have been detected in almost all segments of environment due to their extensive use in past, which have shown potential to biomagnified/accumulate in animal tissue, human blood, adipose tissue and breast milk [7]. Since the pesticides are lipid soluble in nature, cumulative accumulation of low of these in the body fat of mammals might pose potential hazards in long run [8]. The city of Bhagalpur (85°59'E longitude and 25°15'N latitude) is located on the southern bank of river Ganga with an elevation of 172 feet from the mean sea level. Bhagalpur, the silk city, receive about 5 mega gallons of untreated waste water per day including effluents from urban settlement, dying industry, leather shoes and slippers manufacturing units, hospital and pathological laboratories etc. Besides, most of the residents in the neighboring villages of Bhagalpur city are farmers and their livelihood depends on their agricultural produce. The objective of research was to carry out a systemic study on identification and quantification of selected pesticides residues in both water and sediment of river Ganga. The results obtained from this study are presented in this paper.

Material and Methods

Analytical procedure

Liquid-liquid extraction followed by gas chromatographic detector [9] and [10] with some modification was used for the determination of pesticide residues. 1 liter of water sample was filtered using 0.45-µm whatman glass fiber filter paper (washed in acetone and dried in oven overnight). To this 50 ml of phosphate buffer was added and pH was maintained to 7. Now mixture of diethyl ether and hexane in the ratio of 1:1 was taken in a 2 liter separating funnel, and 1 liter of water sample mixed with phosphate buffer was added to it and was shaked vigorously. The layers were allowed to settle down and pass through separating funnel and collected. The extraction was repeated thrice and the solution obtained was filtered with a pinch of sodium sulfate. The combined extract were filtered and concentrated in a vacuorotary evaporator. The solution thus obtained was filtered with a pinch of sodium sulphate and made up to 5 ml with hexane.

The samples thus prepared were injected and analyzed. The pesticides standards (99.9%) purity was supplied from Aldrich Chemicals Germany. Using standard samples containing known amounts of pesticides, accuracy of the determinations was routinely checked. All analysis were carried out in duplicate and recoveries of individual pesticides were determined through spiked sample method, which were found between 98-99.5%.Recovery correction factors were applied to the final results. The results obtained for the various water samples are presented in Tables 3 and Fig. 4.

Study area

The study area included the Gangetic stretch in Bhagalpur starting from Champanala Nathnagar to Burning ghat Barari. (Fig.1 & 2)

A total of three sampling locations were selected on the Southern bank of Jamania river channel and Ganga depending upon the presumed water quality and extent of pollution. They were Champanala (site I, upstream), Mond ghat (site II, midstream) and Burning ghat (Site III, downstream).

Sample Collection

The water sample of the river was collected in sterilized plastic container. Duplicate samples for pesticide measurement were collected from each sampling location. The containers were carefully filled just to overflowing, without passing air bubbles through sample or trapping air bubbles in sealed container. Preparation of the containers included washing with detergent rinsing with tap water and air-drying. The collected samples were preserved in an icebox and transported to laboratory as early as possible for analysis. After transportation to the laboratory, samples were stored at -20°C and extraction was normally done within 48 hours. Sampling was carried out for two years on seasonal basis for the period of January 2006 to December 2007.Samples were analyzed in Pesticide and Toxicology Laboratory, Department of Zoology, Delhi University, Delhi.

Chemicals and instrumentation

The solvents used for extraction i.e. diethyl ether and hexane were obtained from Merck. Standards of insecticides and their metabolites of 99 percent purity were obtained from Aldrich Chemicals Germany. GC- ECD Condition. The pesticide residues were analyzed by gas chromatography (GC) supported by electron capture detector (ECD). The laboratory used Varian GLC with ECD Model No. 6800. Nitrogen was used as the carrier gas and makeup gas and the injection technique was in the split mode .The GC conditions used are furnished in Table 1. The retention times obtained for the various pesticides residues are given in Table 2 and Fig.3.

Results and Discussion

It is well known that most of the applied pesticides are subject to many transport and conversion products. Thus they do not remain at their target site but often enter aquatic environment via soil percolation, air drift or surface runoff affecting abundance and diversity of non-target species producing complex effect on the ecosystems and altering tropics interactions [11].

The results of the analysis of the water samples from river Ganga at Bhagalpur have shown the presence of both organochlorine and organophosphate pesticides. The compounds detected were Lindane, Methyl-parathion, Endosulfan (the isomers alpha and beta endosulfan), and DDT (the isomers orthopara and para-para DDT).

Concentration of organochlorine and organophosphate pesticides (Lindane, Endosulfan and its isomers and DDT and its metabolites) in water of the Ganga River, during different season of study period (2006-07) are summarized in Table 3 and Fig. 4. In Ganga River, no seasonal as well as site trend was observed in the distribution of pesticides. It was invariably higher at site II in river water.

It has also been reported that though DDT is banned but is a part of many organochlorine insecticides, which are not banned. The presence of DDT in high concentration might be attributed to slow degradation of DDT resulting in environmental persistence [12].

Lindane is an organochlorine also known as BHC and HCH. The solubility of lindane in water is 10 mgl^{-1} and reported half-life is of 18 hours. The concentration of lindane in river water was low as compared to other pesticides studied. The concentration of lindane in river water ranged from ND to 74.04 ngl^{-1}. The findings of the present study for river water are in accordance with the findings of [13], [14], [15] and [16]. The order of the concentration of lindane at different sites in river was: site II > site I = site III. Methyl parathion is an organophosphate and is used extensively in agriculture and pest control programs. It has a solubility of 24 mgl^{-1} with a half-life of 10 days to 2 months. Methyl parathion was absent in Ganga river water during the entire study period. The present result is in accordance with the results of [13], [14], and [17] who reported either absence of methyl parathion in river water or present in very low concentration. Methyl parathion being an organophosphate is very toxic for mammals as well as aquatic organisms. In mammals, methyl parathion is a nerve toxin and damages the central nervous system where as in fishes it causes deformation. Endosulfan is an organochlorine and highly toxic pesticide in EPA toxicity. The solubility of endosulfan is 0.3 mgl^{-1} with a half-life of 5 weeks in water but β isomer has longer half-life i.e. 150 days under neutral conditions. Though endosulfan is banned in many countries but it is extensively used in India, especially in these areas with a trade name of *Endocel excel, Endofil 45, Endum* etc. In the present study, T. Endosulfan ranged from BDL to 739 ngl^{-1} in river sediments. High concentration of endosulfan was present in water of the river. α- Endosulfan was higher in water samples, the concentration varying from BDL to 739 ngl^{-1} whereas β Endosulfan was in the range of ND to 157.30 ngl^{-1} in river water. The high concentration of α Endosulfan in water than β Endosulfan in river might be explained as α and β Endosulfan are conformational isomers and can be interconnected without breaking bonds. T-Endosulfan was maximum at site II. This might be due to extensive use of these pesticides in agricultural activities on the riverbanks of site II, and being an organochlorine it has also a tendency to accumulate in the river sediments. The finding of the present study is in accordance with the findings of [15], [18], [19], [16] etc in river water. A local survey of the market of this area reveals that endosulfan is extensively used by the farmers of this region, which might be a reason for presence of endosulfan in such a high concentration. Endosulfan is one of the most toxic pesticides, responsible for many fatal pesticides poisoning incidents. Endosulfan is a xenoestrogen an endocrine disruptor, causing reproductive and developmental damage in animals and humans. It is a neurotoxic in insects and mammals. Endosulfan is highly toxic for aquatic organisms and has bioaccumulating effect especially in fish. The order of concentration of T. Endosulfan in river water was: site II > site I > site III. DDT is an organochlorine insecticide nearly insoluble in water with a half-life of 2–15 years and is immobile in most soils. It is a persistent organic pollutant. DDT along with its isomer o, p' and p, p'-DDT were present and o, p'-DDT concentration was relatively higher than p, p'-DDT in water of river Ganga. This might be attributed to easy metabolization of DDT to its metabolites, which are more stable and persistent than parent molecules of DDT [20]. DDT was present at all the sites in river water at sites III during the study period. In the present study, the T-DDT content ranged from ND to 489 ngl^{-1} in river water. It also indicates about different sources of contamination as well as slow degradation resulting in environmental persistence of these compounds [12]. The present result, with reference to river water, is in accordance with the findings of [14], [21], [22], [15], [23], [19], [20], and [16]. The present result indicates that there is predominant use of DDT in the study area, which as agricultural runoff or surface run off contributes to a heavy load of pesticide pollution in water [24].

Some physico-chemical parameters viz. pH has direct influence on the solubility of these pesticides in river water of Ganga.

The discharge of agro-chemicals from flood plains, agricultural fields through agricultural run off might have contributed to the elevated pesticide concentration at site II. The increased concentration of DDT at site III in water of river might be due to discharge of medical wastes from JLNM College Hospitals, which is channelized directly in to the river near Kali Ghat. Hospitals use DDT for public health activities.

DDT and its metabolic products magnify through the food chain and stored mainly in the body fat. DDT is classified as "moderately toxic" by *US National Toxicological Program* and "moderately hazardous" by WHO. DDT is highly toxic to aquatic life especially fishes as it can bioaccumalate leading to long-term exposure to high concentrations. In human beings, the higher concentration of DDT leads to neuropsychological and psychiatric symptoms. The order of concentration of T-DDT obtained in river water was: site I > site III, whereas T-DDT was either not detected or below detection limit at site II during the study period.

From the above discussion, it is obvious that the order of the concentration of these pesticides in water of river Ganga varied from site to site, and from one location to another location, with vegetable growing areas having higher concentrations of endosulfan and lindane (site II), where as location near maize growing areas contaminated with DDT and its metabolites (site III). At many sites higher concentration of these pesticides were found, indicating that they may present a hazard for riverine ecosystem. The analysis of the results revealed that neither temporal, nor spatial distribution pattern was found, perhaps due to the presence of multiple and aleatory sources. Ganga River receives intermittent inputs of organochlorine and organophosphate pesticides along with their isomers, which are the main contributors to the pesticide pollution of the Ganga River. The presence of DDT metabolites and BHC in the river indicates continuous use of DDT in the catchments. It is noted that though these pesticides are banned in other countries but DDT and BHC are in restricted use in India [25].

Thus we can say that a using pesticide effectively while maintaing water quality presents an important challenge and is need of the hour. As citizens, we must recognize the significant role of pesticides in maintaining a high quality of life. We must acknowledge that the effective production of food and fiber relies on pesticides to control weeds, insects and plant diseases that interfere with the growth, harvest and marketability of crops, and also acknowledge the importance of pesticides in controlling pests in our homes, restaurants, hospitals, parks, ornamental plantings, golf course etc. but at the same time we must be aware that pesticides application can affect water quality of both surface and ground water sources. Human and environmental health may be threatened when excessive concentration of pesticides enters surface or ground water.

Conclusion

Thus we can conclude that the Ganga River gets seriously polluted due to discharge of toxic heavy metals and pesticides from the point and non point sources. As these toxic substances do not degrade, they remain persistent in the environment, and also have the ability to bioaccumulate in the food chain, which might pose potential hazards in long run. Although the toxicity of these elements have been discussed and could itself become a research topic.

The present study was first known analysis of Organochlorine (OCPs) and Organophosphates (OPPs) pesticides distribution in water of river Ganga. The water of river Ganga is polluted by these OCPs and OPPs. A relatively high level of these compounds was observed in most study areas. Most of the organochlorines pesticides found in this study were officially banned, but they were still detected in river water as well as sediments in this area. Regular monitoring and strict law enforcement is needed to develop a strategy to manage the environmental hazards due to these elements and to improve environmental protection of this area. Further it is also suggested that in the present study only 4 pesticides along with the isomers of DDT and Endosulfan was selected for the study but the work of various author on Ganga reveals that there exists a variety of organochlorines and organophosphate pesticide residue and various other toxic trace metals in the water as well as sediment of Ganga river, which have potential health risks to drinking water consumers and organism in Ganga river basin. Further work is needed to determine the bioaccumulation of these toxic elements in the food web and the associated risks to the ecosystem and human health.

Acknowledgement

The authors are thankful to Department of Zoology, University of Delhi and ICAR, Pusa road New Delhi for providing laboratory as well as library facilities. Valuable scientific inputs by Dr. D.K.Singh and Dr. Irani Mukherjee are gratefully acknowledged.

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Table-1: Column specification and operating condition of Gas Chromatograph.

Detector	**Electron Capture Detector (ECD)**
Column specification	30m x 0.25m x 0.25µ Cpsil – 5 – CB
Injection port temperature	280°C
Detector temperature	300°C
Column Programming	170°C hold for 2 min @ 3°C/min 210°C hold for 0 min @ 30°C/min 270°C hold for 2 min Total retention time 20.33 min
Carrier gas	N_2 flow
Split ratio	9 : 1
Carrier gas flow rate	2 ml/min
Back up flow	27 ml/min
Injection volume	2 µl
	Total volume of sample Water – 2 ml. Soil – 5 ml.

Table 2: Retention Time for the pesticides analyzed by the Gas Chromatograph

	Pesticides	**Retention Time** (R.T.) (in min.)
1	Lindane	3.53
2	Methyl-Parathion	4.64
3	α-Endosulfan	8.77
4	β-Endosulfan	10.57
5	o, p' - DDT	11.72
6	p, p' - DDT	13.18

Table 3: Seasonal Pesticides concentration in Ganga river water at Champanala (Site I), Mond ghat (Site II) and Burning ghat (Site III) 2006-2007 at Bhagalpur.

Sites	**Seasons**	**Lindane**	**Methyl Parathion**	**α-Endo sulfan**	**β-Endo sulfan**	**T-Endo Sulfan**	**o,p'-DDT**	**p,p'-DDT**	**T-DDT**
Sites I	Summer	BDL	ND	168.09	ND	168.09	ND	ND	ND
	Monsoon	BDL	ND	BDL	BDL	BDL	78.22	ND	78.22
	Winter	BDL	ND	108.02	BDL	108.02	56.78	ND	56.78
	Summer	BDL	ND	130	ND	130	ND	ND	ND
	Monsoon	ND	ND	BDL	BDL	BDL	489.00	ND	489.0
	Winter	ND	ND	140.08	BDL	140.08	489.00	ND	489.0
Sites II	Summer	BDL	ND	BDL	125.09	125.09	ND	ND	ND
	Monsoon	74.04	ND	512.11	BDL	BDL	ND	ND	ND
	Winter	BDL	ND	208.09	ND	208.09	ND	ND	ND
	Summer	ND	ND	BDL	157.30	157.30	ND	ND	ND
	Monsoon	48.30	ND	739	ND	739	ND	ND	ND
	Winter	BDL	ND	739	ND	739	ND	ND	ND
Sites III	Summer	BDL	ND	145	ND	145	ND	ND	145
	Monsoon	ND	ND	BDL	BDL	BDL	100.0	112.0	212.0
	Winter	ND	ND	129.08	BDL	129.08	125.0	BDL	125
	Summer	BDL	ND	BDL	BDL	BDL	ND	ND	ND
	Monsoon	BDL	ND	BDL	BDL	BDL	BDL	ND	ND
	Winter	BDL	ND	BDL	BDL	BDL	BDL	ND	ND

BDL = Below detection limit, ND = Not detected, Detectable limit: Lindane = 1.00, Me. Parathion = 10.00, α-Endosulfan and β-Endosulfan = 10.00, o, p'-DDT and p, p'-DDT = 1.00. All values in nanogram/ litre = ngl^{-1}

(Analysis done at Pesticide in *Toxicology Laboratory*, Dept. of Zoology, Delhi Univ., Delhi, using *Varian GLC with ECD, Model No. 6800*).

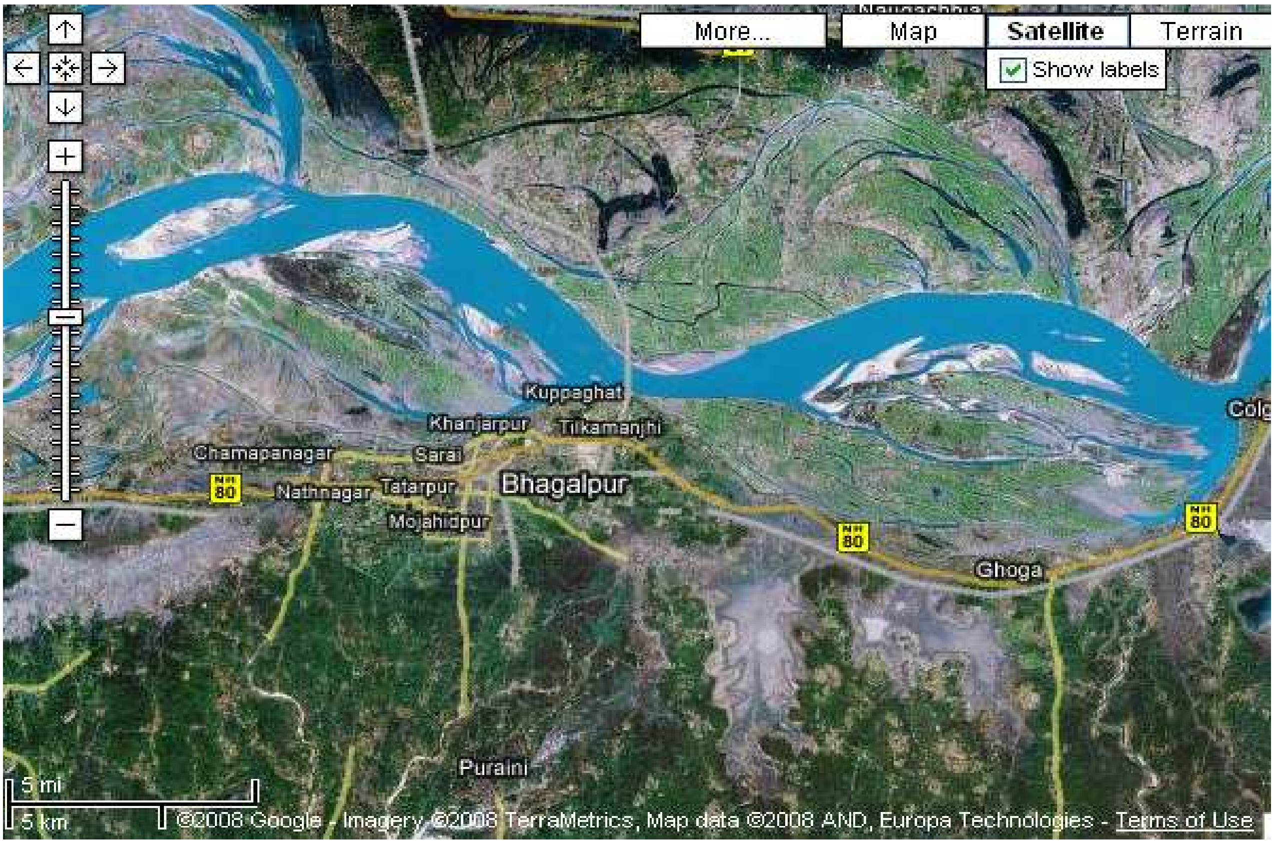

Figure 1 : Satellite Imagery of River Ganga in Nathnagar-Bhagalpur Stretch (Bihar)

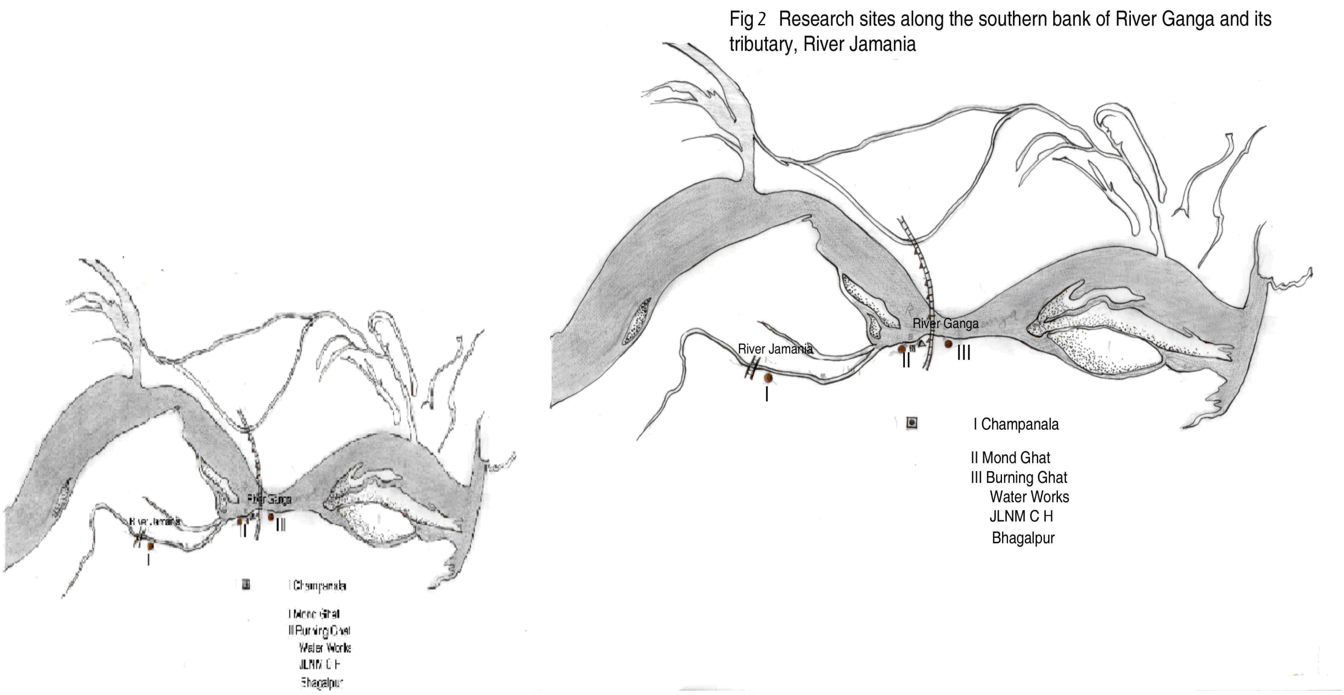

Figure 2: Research Site along the southern bank of River Ganga and its tributary, River Jamania

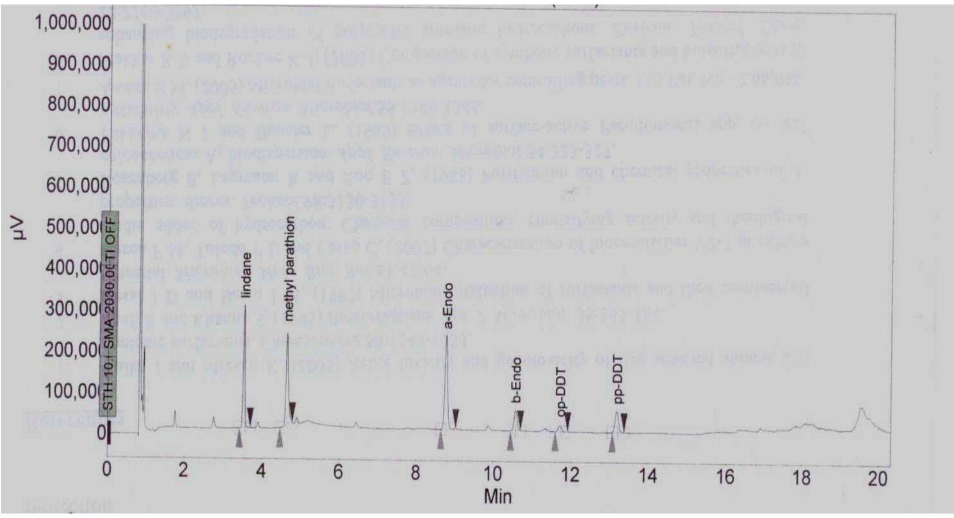

Fig. 3 Chromatogram showing retention time for six different Pesticides (Organochlorine and Organophosphate)

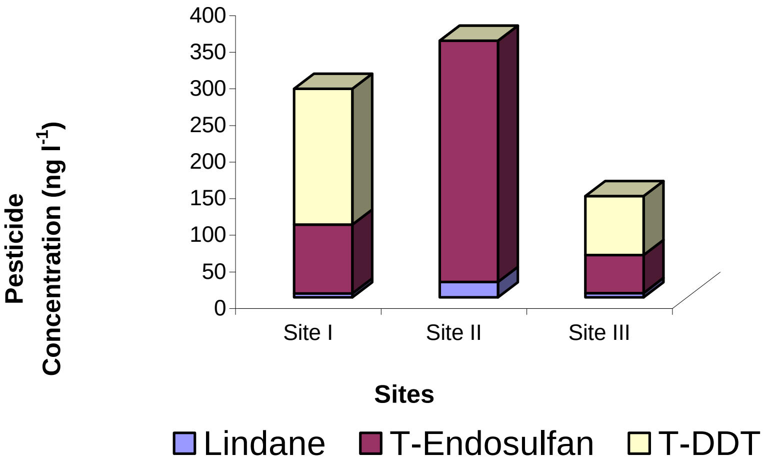

Figure 4: Range of Concentration of Pesticide at Various Sites in Water of River Ganga (2006-2007).