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Detection of Organochlorine and Organophosphorus Pesticides Residues in Water Samples of Daudkandi Thana in Comilla District in Bangladesh

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Abstract: *This study was undertaken to investigate the extent of Organochlorine and Organophosphorous pesticides residues in water samples from some paddy fields ditches, ponds and tubewells in Daudkandi Thana of Comilla district in Bangladesh by using High Performance Liquid Chromatography (HPLC). Among 25 samples, Organophosphorus (Malathion) pesticide was found in eleven samples while Organocholorine was absent. The level of the pesticide residues (Malathion) were found in the range of 1 to 82 ppm, which were above the maximum acceptable levels of total and individual pesticide contamination.*

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I. Introduction

Bangladesh is an agricultural country and agriculture governed its economy. To support a rapidly large growing population of the country huge production of crops are required and for this reason HYVs rice was introduced and have been contributing notably to the self sufficiency in food production but at the cost of environmental pollution as agrochemicals have been used intensively for this purpose[1]. Around 40% of the crop loss is caused by pests and insects which lead to the recurrent use of pesticides[2,3]. Potential hazardous pesticide use is now an integral part of intensive crop production[4]. Use of pesticide in Bangladesh increased at a rate of 10.0% per year and pesticide overuse was evident at national and farm-level which indicated a deplorable situation[5]. Concerns over the use of pesticides in agriculture include contamination of surface water, ground water, soils and food and consequent impacts on human, fish and wildlife[1]. Pesticides can contaminate surface water through various pathways including agricultural run-off, spray drifts, atmospheric fall out, direct spills or accidental discharge[6]. Farmers often overuse hazardous pesticides such as organophosphates and organochlorines which possibly contaminate groundwater or potable water in various ways[1]. Organochlorine and organophosphate pesticides are now of great concern because of their toxicity, tendency for long-range transport and bioaccumulation in food chain posing threat to human health[6]. Although many pesticides including some organochlorine and organophosphate pesticides are banned in several countries of world still they are being used in Bangladesh because of their easy availability, low price, wide spectrum capability and lack of the farmers about those pesticides. So, regular monitoring of water samples is needed to take effective measures for the control of use of pesticides and to save the surface water from contamination.

Daudkandi thana of Comilla district is famous for crop production like rice, wheat, potato and various vegetables. Consequently, pesticides have been used legally or illegally for various purposes. But adequate information is not available on the pesticide residues in the different water samples of Daudkandi. Moreover, there is no known works have yet been done for the determination of pesticides levels in water samples at Daudkandi to investigate the effects of random pesticides use in agricultural fields. Therefore, Daudkandi Thana of Comilla district has undertaken as the study area. The present study was undertaken to assess the contamination level of water samples by organochlorine and organophosphorus pesticide residues from the different sources of the study area and to propose a recommendation about the pesticide use of the region.

II. Material And Methods

The experiment was conducted to study on the monitoring of pesticide residues from three different sources of ditch water, pond water and tubewell water from 5 unions of Daudkandi thana of Comilla district.

Study Design: Random sampling.

Study Location: Comilla is a major district located near the capital of the country Bangladesh. Comilla District with an area of 3,085.17 sq km, is bounded by Brahmanbaria and Narayanganj districts on the north, Noakhali and Feni districts on the south, Tripura (state of India) on the east, Munshiganj and Chandpur districts on the west (fig. 1). Annual average temperature maximum 34.3^0 C, minimum 12.7^0 C; annual rainfall of 2551 mm. Main rivers are Meghna, Gumti and Dakatia. Daudkandi thana of the comilla district is famous for rice and other crop production. Lands are mostly used for the production of rice, jute, wheat, potato, mustard and other vegetables. This Daudkandi thana is compromising five unions: Daudkandi, Goalmari, Sundalpur, Baropara and Gouripara (fig.2).

Fig 1. Map of Comilla

Fig 2. Map of Daudkandi

Study Duration: October 2015 to December 2015.
Sample size: A total of 50 samples.

Procedure methodology

Water sampling and analysis: Samples were collected from 10 villages of five unions *i.e.* Daudkandi, Goalmari, Sundalpur, Baropara, Gouripur. Water samples from respective spots and types were collected and brought to the Agrochemical and Environment Research Division (AERD), Institute of Food and Radiation Biology (IFRB), Atomic Energy Research Establishment, Savar, Bangladesh for subsequent processing and analysis.

Sample preparation: 250 mL of water sample and 100 mL double distilled-hexane (DD-hexane) as solvent were taken into a separating funnel and gently shaken. After 10-20 minutes the lower aqueous layer and upper thick solvent layer of hexane with pesticide residues were collected in separate conical flasks. Re-extraction of the aqueous layer was done for two times by adding 50 mL solvent (DD-hexane) and then the solvent layer (upper) was collected. Combined extract was collected with Na_2SO_4 (anhydrous) for removing water (if any). The collected extract was then concentrated using a rotary vacuum evaporator, and the extract was transferred into a cleaned and rinsed vial by rinsing with DD hexane for three times and making the volume 7 mL. In case of color formation, clean-up of the extract was done over florisil and eluted with 2% diethyl ether n-hexane. Again, the extract was evaporated at 40^0C by vacuum rotary and transferred into vials. The extracts were then evaporated by nitrogen gas and dried completely. Final volume was made adding 1 mL of acetonitrile prior to injection into HPLC for pesticide detection. Then the samples were analyzed with HPLC (High Performance Liquid Chromatography)[7].

Statistical analysis: All the data obtained from the experiment were analyzed by using SPSS 16.0. From the analysis no significance difference of concentration level of pond, ditches and tube well was found (ANOVA, LSD, $p<0.05$)

III. Result

By High Performance Liquid Chromatography (HPLC) all the tests of water samples was done for organochlorine and organophosphorus pesticide residues. Results have been presented in the table 1and 2. None of the water samples showed the presence of DDT, DDD or DDE residues. Diazinon and Chloropyrifos residues found only in one sample whereas Malathion found in eleven samples and most of them were from ditch and pond water. When all the collected water samples were analyzed in the chromatograph, peaks of any water

samples tested did not resemble to any of the peaks of the mixed standard chromatogram of Organochlorine pesticides. During the analyses some other small peaks were also observed. It may be due to the presence of unknown contaminants; this contamination may arise from the injection syringe, vial or any other ways. Residues from the pesticides leach into the water, which are harmful to anything in the water

Table 1: Results for Organochlorine pesticide residues in µg/L in water samples from Daudkandi thana of Comilla district, Bangladesh

Sl No	Sample No	DDT	DDD	DDE
1	1-D	ND	ND	ND
2	1-P	ND	ND	ND
3	1-T	ND	ND	ND
4	2-D	ND	ND	ND
5	2-P	ND	ND	ND
6	2-T	ND	ND	ND
7	3-D	ND	ND	ND
8	3-P	ND	ND	ND
9	3-T	ND	ND	ND
10	4-D	ND	ND	ND
11	4-P	ND	ND	ND
12	4-T	ND	ND	ND
13	5-D	ND	ND	ND
14	5-P	ND	ND	ND
15	5-T	ND	ND	ND
16	6-D	ND	ND	ND
17	6-P	ND	ND	ND
18	7-D	ND	ND	ND
19	7-P	ND	ND	ND
20	8-D	ND	ND	ND
21	8-P	ND	ND	ND
22	9-D	ND	ND	ND
23	9-P	ND	ND	ND
24	10-D	ND	ND	ND
25	10-P	ND	ND	ND

D= Ditch water, P= Pond water, T= Tubewell water, ND= Not Detected

Table 2: Results for Organphosphorus pesticide residues in µg/L in water samples from Daudkandi thana of Comilla district, Bangladesh

Sl No	Sample No	Malathion	Diazinon	Chloropyrifos
1	1-D	82	ND	ND
2	1-P	ND	ND	ND
3	1-T	ND	ND	ND
4	2-D	ND	ND	ND
5	2-P	62	ND	ND
6	2-T	ND	ND	ND
7	3-D	7	ND	ND
8	3-P	48	0.651	0.031
9	3-T	ND	ND	ND
10	4-D	6	ND	ND
11	4-P	54	ND	ND
12	4-T	ND	ND	ND
13	5-D	3	ND	ND
14	5-P	7	ND	ND
15	5-T	ND	ND	ND
16	6-D	ND	ND	ND
17	6-P	8	ND	ND
18	7-D	ND	ND	ND
19	7-P	ND	ND	ND
20	8-D	1	ND	ND
21	8-P	ND	ND	ND
22	9-D	1	ND	ND
23	9-P	ND	ND	ND
24	10-D	ND	ND	ND
25	10-P	ND	ND	ND

D=Ditch water, P= Pond water, T= Tubewell water, ND= Not Detected

IV. Discussion

Effects of farm pesticides on water quality in Lake Naivasha, Kenya were studies and no organochlorine or organophosphate residues were detected in the water[8] and in this study no organochlorine residues was detected but organophosphate residues were detected. Organochlorine and organophosphorus pesticide residues in water and sediment from Yala/Nzoia river within lake Victoria basin, Kenya were studies with a focus on seasonal variation and it was found that banned organochlorines were still being used in the catchment and organochlorines such as dieldrin and DDD present in water sample and were found below the detection limit and no organophosphate except diazinon and malathion were detected and the residues were found in the sediment sample of the river mouth and the concentration was below 0.03µg/kg at Yala and 0.01µg/kg Nzoia river[9]. Pesticide residues in water samples from Meherpur region of Bangladesh were investigated for the presence of organophosphorus and carbamate pesticide residues by HPLC where water samples were found to be contaminated with diazinon (0.033 to 0.079 ppm), Chloropyrifos (0.010 to 0.471ppm) and carbofuran[10] and that residue level was within the acceptable range according to WHO standard for water quality[11]. Presence of DDT, DDE and dieldrin were detected from the irrigated crop fields in Gaibandha, Bangladesh and in most cases concentration level was below WHO guidelines but on the other hand, water samples from Begumganj were found to contain DDT level at 19µ g/L which was above WHO guideline value[11,12]. Pesticide residues in water and sediment samples of the river Challawa were detected which were diazinon(0.21-0.61mg/L), chloropyrifos(0.35-0.78mg/L), dieldrin(0.05-0.25mg/L), Aldrin(o.11-0.36mg/L), DDT(0.01-0.09mg/L), DDE(0.01-0.21mg/L) and DDD(0.01 -0.24 mg/L)[13]. The pesticides concentration levels in water samples adjacent to agricultural fields of Savar upazila in Bangladesh by HPLC where chloropyrifos was found in four samples and concentration ranged from 3.27 -9.31µg/L, diazinon was found in one sample (7.86µg/L), Malathion found in 2 samples (23.1 and 59.9 µg/L) and DDT was below detection limit[3]. Another study was conducted at Dhamrai Upazila beside Savar Upazila to identify and quantify organochlorine (DDT, DDE and DDD) and organophosphorus (malathion, diazinon, chloropyrifos) residues in water samples from different sources *i.e.* fish pond, cultivated land and tubewell in winter season and it was found that among 30 water samples, Malathion (organophosphorus pesticide) was present in 7 samples ranging 42.58 to 922.8µg/L and diazinon in one sample (31.5 µg/L) and DDT, DDE, DDD and chloropyrifos were not detected in any sample and it was noted that in most cases, the concentration of the detected residues were above the acceptable level for human body[14]. In this study, the maximum malathion concentration was found 82µg/L and the value was above the acceptable level according to Australian health based guideline value of 70µg/L[15] though most of the values are below the maximum acceptable level. Similar results for malathion were obtained from the water samples of Dhamrai[3, 16,17]. Chloropyrifos was not detected in the current study but it was found in the water samples from Rangpur district of Bangladesh at the range of 0.477 to 1.189µg/L[16]. Diazinon found in the current study was much below the acceptable limit of 4 µg/L[15] but water samples of Dhamrai Upazila had higher limit[14] and similarly 32.8-79µg/L of diazinon were found in Meherpur region[10] whereas 0.9µg/L concentration was found in Savar Uapazila[16] and 0.027µg/L concentration in Manikgonj in Bangladesh[18]. As mentioned earlier DDT, DDE and DDD were not found in any sample in the current study which is similar to the water samples investigated in Dhamrai Upazila[14] but concentration of 19.6 µg/L of DDT in Begumganj[19], Bangladesh and concentration ranging from 0.133 to 8.29 µg/L was reported in several districts of Bangladesh and DDE was detected in the water samples of Burichang of Bangladesh with the level of concentration of 4.06µg/L[20].

Pesticides not only kill target pests but some other non-target animals as well. In pesticide contaminated water fish population often die or leads to a decrease in fish yields and losses of fishing grounds. The death of birds and fish due to pesticides is most common, which has led to bans of certain pesticides in some places[21]. Malathion is used in wide scale in rice field in Bangladesh. Most of the farmers in Bangladesh applied Malathion at the higher dose than the recommended dose of 16.8 kg/ha[22]. The rate of disappearance of Malathion from soil has been reported to be 75 to 100% in one week[23]. The samples in the experiment might not show the exact concentration of residue as the samples were collected during late autumn season which was the season of rice harvesting. Generally pesticides were applied in the crop field 1 to 2 months prior to harvesting and thus the pesticide residue might degrade that time. Some samples were collected from vegetables fields, where pesticides were used 15-20 days ago. During that period there was no rain to run off the pesticide to nearby water body. As Organophosphorus degrades quickly in between 1-2 weeks[24] and as a result those pesticides might not be identified. It may be noted that Malathion is a nonsystemic, wide spectrum aliphatic organophosphate insecticide widely used for both domestic and commercial agricultural purposes. Malathion is only found in individuals that have used or taken this drug. It is a wide spectrum aliphatic organophosphate insecticide widely used for both domestic and commercial agricultural purposes. It inhibits acetylcholinesterase activity of most eukaryotes. Malathion is toxic to aquatic organisms, but has a relatively low toxicity for birds and mammals. The major metabolites of malathion are mono- and di-carboxylic acid derivatives, and malaoxon is a minor metabolite. However, it is malaoxon that is the strongest cholinesterase inhibitor.

Cholinesterases catalyze the hydrolysis of the neurotransmitter acetylcholine into choline and acetic acid, a reaction necessary to allow a cholinergic neuron to return to its resting state after activation. Because of its essential function, chemicals that interfere with the action of cholinesterase are potent neurotoxins, causing muscle spasms and ultimately death.

V. Conclusion

This is very preliminary research which gives a baseline data of pesticide residue levels in the water samples of Daudkandi thana of Comilla district. From the survey it can be said that there is evidence to conclude that organochlorine pesticides residues (DDT and its derivatives) were absent in the water samples analyzed. So, the farmers of Daudkandi thana may be not using organochlorine as pesticide but some organophosphorus pesticides such as Malathion were being used by the farmers as it was found in eleven samples.

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