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ORIGINAL ARTICLE

Persistent Organic Pollutants and Pesticide Residues in Seasonal Waters of Rural Bangladesh

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Abstract

Pesticide use in Bangladesh is often excessive and unregulated. Despite their worldwide ban, Persistent Organic Pollutants (POPs) and other harmful organochlorine pesticides are widely being used. No studies have been done in Bangladesh on the presence of POPs in water from and around agricultural fields. This study detected POPs such as HCB, O, P`– DDT, DDD, 4, 4`-DDT, α-HCH, β-HCH and δ-HCH in water samples. The samples were analyzed using standard test methods. The tests indicated that at least one of the seven pesticides was in every sample collected. The concentration of analytes ranged from 121.793×10^{-2} μgL^{-1} for 4, 4`-DDT to 0.014×10^{-2} μgL^{-1} for HCB. 13 of the 15 samples had at least one of the three isomers of DDT in them. δ-HCH was present in all the samples. The presence of POPs in every sample indicates continued use of harmful pesticides in the country.

Keywords: *Agriculture, Pesticides, Bangladesh, POPs.*

I. INTRODUCTION

The economy of Bangladesh is mainly based on agriculture. The country has a tropical climate and the major crops are rice, wheat, jute, potato, sugarcane, vegetables and tea [1]. The hot and humid weather of the region is highly favorable for the growth of pests. Also, new varieties of rice and other crops are coming into cultivation almost every year due to fruitful researches being carried out by International Rice Research Institute (IRRI) and other national agricultural research organizations. Some of these varieties are especially prone to attack of pests. The farmers, most of whom are not very well-to-do and not well-educated, go for any kind of pesticides that come their way easily and cheaply [2, 3].

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There is thus widespread use of pesticides, herbicides and chemical fertilizers in the country. There have been growing concerns about the environmental impact of excessive use of pesticides [4, 5]. Among the pesticides being used, some of the organochlorine pesticides are known to be harmful for human health and the environment [6-10]. Some of these organochlorine pesticides have been banned worldwide due to their persistence and bioaccumulative nature [11, 12]. One such group of organochlorine pesticides has been classified as Persistent Organic Pollutants (POPs). In May 1995, United Nations Environment Program (UNEP) has recognized POPs as an imminent danger for human health and made a short list of twelve POPs, commonly known as the dirty dozen [13]. In 2009, nine additional chemicals were added to the list [14]. POPs have been shown to resist degradation by physical, chemical, biological, and photolytic processes [11]. Various studies all over the world have shown the presence of a large variety of POPs in different matrices, such as water, air, fish, soil, solid wastes, human tissues, etc. [15-20]. POPs' persistence and semi-volatile nature enables them to be transported by air, water or other means to remote regions where they have never been used or released. After their release into the environment, they travel multiple cycles of evaporation, transportation by air and condensation [21]. POPs' health effects include cancer, birth defects and reproductive discrepancies, behavioral problems, immunological and neurological disorders in

both humans and animal species [8, 9, 22-25]. Despite their worldwide ban, POPs are widely used in developing countries, especially South Asian countries [6]. Greenpeace has found DDT, dieldrin, heptachlor along with other harmful pesticides openly sold in the markets of Bangladesh [26]. According to a study conducted by a local NGO named Environment and Social Development Organization (ESDO), POPs are still widely used and released to the environment across the country [6]. There have been DDT stockpiles in different districts which are contaminating the environment as the stores are not well managed. The major concentration of these hotspots in Bangladesh has been found in Dhaka city and the surrounding areas namely Gazipur, Narshingdi, Narayanganj, and also in Chittagong, Khulna, Sylhet and Barishal [1].

Some recent studies have found DDT in lake, pond and hand tube well water of the Dhaka metropolitan city area and also in dry fish and vegetables like cauliflowers [27, 28].

Though they are generally considered to be persistent, in some cases, POPs do degrade in the environment. When breakdown does occur, it creates chemicals that are also hazardous [29]. Commercial DDT is actually a mixture of several closely related compounds. The major component (77%) is the 4.4` isomer. The O,P' isomer is also present in significant amounts (15%). Dichlorodiphenyldichloroethylene (DDE) and dichlorodiphenyldichloroethane (DDD) make up the balance. DDE and DDD are the major isomers and breakdown products of DDT in the environment [12].

Another POP, hexachlorobenzene (HCB), or perchlorobenzene, is a fungicide used as a seed treatment, especially on wheat to control the fungal disease [25]. HCB is also formed as a byproduct during production of a variety of chlorinated compounds. HCB is an animal carcinogen and is considered to be a probable human carcinogen. HCB is very toxic to aquatic organisms and may cause long-term adverse effects in the aquatic environment [30].

Hexachlorocyclohexane (HCH) is also used extensively as an insecticide. HCH is an isomeric mixture that contains mainly five forms of HCH. They are alpha-hexachlorocyclohexane (α-HCH), beta-hexachlorocyclohexane (β-HCH), gamma-hexachlorocyclohexane (γ-HCH), delta-hexachlorocyclohexane (δ-HCH) and epsilon-hexachlorocyclohexane (ε-HCH). α-HCH and β-HCH are byproducts of the production of the insecticide lindane (γ-HCH). It is typically still contained in commercial grade lindane. δ-HCH is not part of POPs yet but it is being considered to be included in the list. Like other POPs, δ-HCH is persistent and bioaccumulative. It is no longer produced or sold for domestic use, but its significant presence in environment is of concern [30]. Isomers of HCH act as stimulants and central nervous depressants. Due to its lipophilicity and persistence, isomers of HCH frequently accumulate in human adipose and breast tissues [31]. Human studies have shown that exposure to HCHs is linked to cancer, Parkinson's, Alzheimer's, reproductive and fertility disruption [31-33].

This paper focuses on the quantitative detection of six different POPs (HCB, O, P`–DDT, DDD, 4, 4`-DDT, and α-HCH and β-HCH) and δ-HCH in water samples from and around agricultural fields. The concentrations of POPs were determined using GC-ECD, the standard method for detecting organochlorine pesticides in water [34].

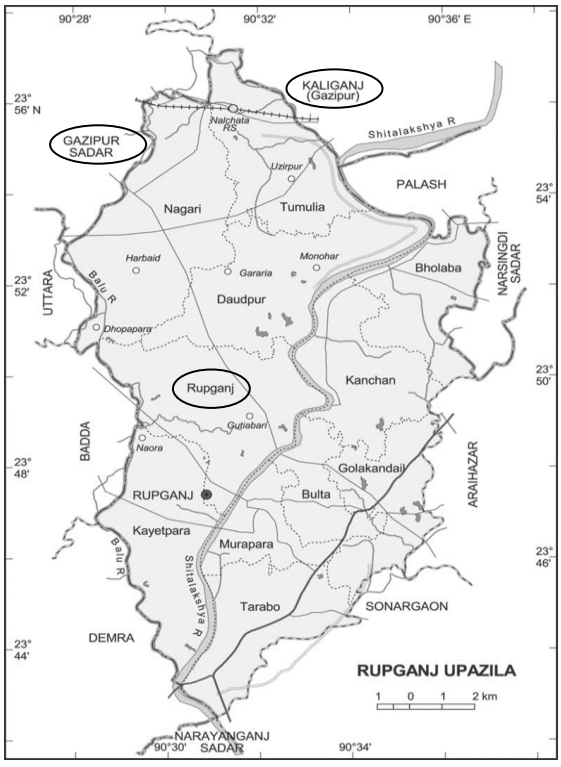

Fig. 1. Sample sites

II. Materials and Methods

A. Sampling area

For the convenience of collection of samples, the sites were chosen near the capital, Dhaka. The selected sample sites were agricultural lands located in Gazipur, Rupganj and Kaliganj. Rupganj and Kaliganj are in the district of Narayanganj. Dhaka has the district of Gazipur

at its north side and that of Narayanganj at the south side. Both Gazipur and Narayanganj districts have been identified as the major hotspots with DDT stockpiles [1]. The sample sites have been indicated in Fig. 1 [35].

B. Sampling

The procedures for sampling, extraction and clean-up were followed according to established methods [36]. Fifteen water samples from and around rice, jute and sugarcane fields were collected immediately after the monsoon season. The samples were collected in this season because the farmers apply pesticides usually during and immediately before the monsoon season. Out of the total 15 samples, 9 were collected from the ditches adjacent to crop fields. The remaining 6 samples were collected from the stagnant water present inside the crop fields. Samples were collected directly into pre-cleaned plastic sample bottles. Care was taken not to disturb the surface of the soil layer while collecting the samples. The water samples were not filtered to separate suspended particles because the suspended particles, especially fat particles, could contain POPs in them. Samples were kept below 4°C until analysis. The pesticide standards were purchased from Sigma-Aldrich with the purity of 99.8%. All other chemicals, solvents and reagents used in this study were of analytical grade.

500 mL of each water sample was taken in a separating funnel and 50g of sodium chloride was added. The content was extracted three times with 50 mL of n-hexane. 30 g of anhydrous sodium sulphate was added to the combined extracts. The extracts were filtered and the water-free organic layer was taken in an evaporation flask. The volume was carefully reduced to about 0.5 – 1.0 mL by evaporation. The sample was cleaned up with 2 mL of 95 - 97% pure sulphuric acid saturated with cyclohexane. The mixture was left for separation and the upper phase was taken for analysis in a gas chromatograph.

C. Analytical methods

Gas chromatographic analyses were carried out on a Shimadzu Model 2010 Gas Chromatograph equipped with a ^{63}Ni electron capture detector with ultrapure nitrogen gas (99.9%) as the mobile phase.

Chromatographic determinations were carried out using a 30 m x 0.53 mm x 3.00 µm dimethyl siloxane capillary column (DB-624) manufactured by J & W. The operating conditions were as follows: injection mode was splitless; injection port temperature was 210.0°C; detector temperature was 300.0°C; column temperature was raised from 240.0°C to 260.0°C. Peak areas were used as the basis for quantification. Standard solutions of HCB, O, P`– DDT, DDD, 4, 4`-DDT, and α-HCH, β-HCH, and δ-HCH were made from double distilled, deionized water. Each of the standards was run thrice in the gas chromatograph to check whether the retention time was reproducible. After measuring the retention times of each standard, a mixture of standards was analyzed to verify whether all the retention times remained the same. In order to check the reliability of the experimental results, samples were spiked with 10 μgL^{-1} of the mixed standards and the resultant peak areas were compared with the calculated values of the analytes. The concentration in samples was expressed in μgL^{-1}.

III. Results and Discussion

Table 1 shows the areas from where samples were collected, the kind of crops that were being cultivated and also indicates whether the samples were collected from an adjacent ditch or from the stagnant water present inside the crop fields.

Table 1. Sample site

Area	No.	Kind of crops	Source
Rupganj	1	Jute, Sugarcane	Adjacent ditch
	2	Jute, Sugarcane	Adjacent ditch
	3	Rice	Adjacent ditch
	8	Rice	Adjacent ditch
	10	Jute, Sugarcane	Adjacent ditch
	11	Rice	Water from field
	12	Sugarcane	Adjacent ditch
	13	Sugarcane	Adjacent ditch
	14	Jute	Adjacent ditch
	15	Sugarcane	Water from field
Kaliganj	6	Jute	Water from field
	7	Jute	Water from field
	9	Jute	Water from field
Gazipur	4	Rice	Adjacent ditch
	5	Rice	Water from field

Note: Each serial number represents a different sample site.

The concentrations of the POPs detected in the samples are shown in Table 2.

Table 2. Concentrations of POPs in Water Samples from Agricultural Fields

No.	Concentration (μgL^{-1} x 100)						
	HCB	O,P`DDT	DDD	4,4`DDT	α-HCH	β-HCH	δ-HCH
1	0.352	ND*	ND	8.655	ND	ND	7.703
2	0.985	43.362	ND	58.496	29.133	3.673	56.662
3	ND	ND	ND	ND	ND	0.775	2.429
4	1.584	ND	5.871	105.113	ND	0.421	49.525
5	10.297	5.000	5.000	10.206	ND	ND	10.117
6	0.530	ND	8.279	112.219	ND	ND	51.790
7	11.010	2.927	3.690	56.816	ND	ND	45.653
8	1.344	ND	ND	65.242	ND	ND	13.781
9	0.767	ND	4.837	91.653	ND	ND	100.386
10	0.191	ND	7.626	47.530	ND	ND	30.567
11	0.371	ND	7.778	121.793	ND	ND	79.486
12	ND	9.345	ND	49.812	ND	ND	24.892
13	ND	ND	ND	2.873	0.295	0.574	3.158
14	0.014	ND	ND	ND	ND	0.018	0.955
15	0.096	0.349	ND	10.868	0.056	ND	8.954

* ND means not detected.

Note. USEPA and World Health Organization limit for HCB in water is 1.0 μgL^{-1} [37], [38].
European Union limit for O,P`DDT in water is 0.1 μgL^{-1} [39].
European Union limit for 4, 4`DDT in water is 0.1 μgL^{-1} [39].
European Union limits for α-HCH in water is 0.1 μgL^{-1} [18].
European Union limits for β-HCH in water is 0.1 μgL^{-1} [18].
European Union limits for δ -HCH in water is 0.1 μgL^{-1} [18].

Since rice, jute and sugarcane are some of the main agricultural crops in Bangladesh, the samples for this study were collected from these fields (Table I). Six of the water samples were collected from stagnant water present inside the crop fields, while nine samples were collected from adjacent ditches. The local people often use water from the ditches for cooking and washing their utensils. Sometimes, they even take bath in these ditches that are deeper. Children also enjoy playing in these waters.

All the samples collected had at least one of the seven organochlorine pesticides present. No sample showed the presence of all seven target pesticides, but sample number 2 showed the presence of six of the seven target pesticides.

Twelve of the fifteen samples were found to contain HCB ranging from 0.014×10^{-2} μgL^{-1} to 11.010×10^{-2} μgL^{-1}. It is known that HCB is used as a fungicide and the presence of HCB in the water samples in and around agricultural fields indicates that HCB is being used.

Thirteen of the fifteen samples were found to contain 4, 4` DDT with concentrations ranging from $2.873\times10^{-2}$ to $121.793\times10^{-2}$ $\mu gL^{-1}$ with three values exceeding the limit set by the European Union [39]. It is known that major component (77%) of the commercial DDT used as pesticide is the 4,4` isomer [10]. It can be speculated from the results that commercial DDT is being used in almost all the sample sites. DDD was found in 7 samples and their concentrations ranged from 3.690×10^{-2} μgL^{-1} to 8.279×10^{-2} μgL^{-1}. O,P` DDT was found in only 5 samples and the concentrations varied from 0.349×10^{-2} μgL^{-1} to 43.362×10^{-2} μgL^{-1}. One of the samples (sample 2) was above the regulatory limit [39]. Almost all the samples had at least one of the three isomers of DDT in them. The presence of these DDT isomers is an indication of the continued use of POPs in agriculture.

All the fifteen samples collected from different areas contained δ-HCH at concentrations ranging from 0.955×10^{-2} μgL^{-1} to 100.386×10^{-2} μgL^{-1}. This study suggests that δ-HCH is being used as an insecticide in

all locations sampled. Nine samples had δ-HCH concentration at a higher value than the standard value set by the European Union [18]. α-HCH and β-HCH were not as common as δ-HCH. α-HCH was found in only 3 samples and β-HCH was found in only 5 samples. α-HCH was found to exceed the limit in sample 2 [18]. α-HCH and β-HCH are byproducts of the production of the insecticide lindane [40].

Although concentration of POPs in most of the samples were within the regulatory limits, it must be emphasized that POPs are inherently unmanageable and they bioaccumulate in living species. Therefore, the acceptable standard for any POP in any sample should ideally be zero [18].

IV. Conclusion

The analyzed results of this study indicate the use of organochlorine pesticides like HCB, DDT, α-HCH, β-HCH, δ-HCH in agriculture of the country. Even though these pesticides are banned in the country, they are still being used. In most of the cases, the samples with higher concentrations were those that were collected from inside the fields. This seems to lend support to our hypothesis that the POPs found in these stagnant waters were coming from the pesticides applied in the fields. Further studies should be carried out in larger and more targeted sample areas with all POPs and other significant organochlorine pesticides.

Since POPs have much higher affinity to fats and sediments compared to water, the amount of POPs in these matrices must be much higher than what have been found in these waters. Worldwide studies have found HCB, DDT and α-HCH, β-HCH, and δ-HCH in human blood, breast milk and adipose tissues, but so far no study of this nature has been carried out in Bangladesh [22], [41], [42]. Various food and agricultural crops can also be analyzed for the presence of POPs in the food chain. As the present study found existence of POPs in surface water, chances of groundwater contamination should also be addressed.

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