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Pesticide residue level in tea ecosystems of Hill and Dooars regions of West Bengal, India

Avhik Bishnu · Kalyan Chakrabarti · Ashis Chakraborty · Tapan Saha

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Abstract In the present study we quantified the residues of organophosphorus (e.g. ethion and chlorpyrifos), organochlorine (e.g. heptachlor, dicofol, α-endosulfan, β-endosulfan, endosulfan sulfate) and synthetic pyrethroid (e.g. cypermethrin and deltamethrin) pesticides in made tea, fresh tea leaves, soils and water bodies from selected tea gardens in the Dooars and Hill regions of West Bengal, India during April and November, 2006. The organophosphorus (OP) pesticide residues were detected in 100% substrate samples of made tea, fresh tea leaves and soil in the Dooars region. In the Hill region, 20% to 40% of the substrate samples contained residues of organophosphorus (OP) pesticides. The organochlorine (OC) pesticide residues were detected in 33% to 100% of the substrate samples, excluding the water bodies in the Dooars region and 0% to 40% in the Hill region. The estimated mean totals of studied pesticides were higher in fresh tea leaves than in made tea and soils. The synthetic pyrethroid (SP) pesticide residues could not be detected in the soils of both the regions and in the water bodies of the Dooars. Sixteen percent and 20% of the made tea samples exceeded the MRL level of chlorpyrifos in Dooars and Hill regions respectively. The residues of heptachlor exceeded the MRL in 33% (April) and 100% (November) in the Dooars and 40% (April) and 20% (November) in the Hill region. Based on the study it was revealed that the residues of banned items like heptachlor and chlorpyrifos in made tea may pose health hazards to the consumers.

Keywords Maximum residue limit · Pesticide residues · Tea

A. Bishnu (✉) · T. Saha
Department of Environment,
Institute of Environmental Studies and Wetland Management,
B4 LA Block, Sector-III, Salt Lake,
Kolkata, West Bengal 700098, India
e-mail: avhikb@yahoo.co.in

K. Chakrabarti
Department of Agricultural Chemistry and Soil Science,
Calcutta University,
35 Ballygunge Circular Road,
Kolkata, West Bengal 700019, India

A. Chakraborty
Department of Agronomy,
Bidhan Chandra Krishi Viswavidyalaya,
Mohanpur, Nadia, West Bengal 741252, India

Introduction

Tea (*Camellia sinensis* L.) is one of the most important cash crops in India due to its tremendous export potentialities. It is grown in the state of West Bengal, an eastern state of India, covering an area of about 1.03 million ha in Darjeeling, Terai and Dooars regions and contributes about 21% of the total tea production of the country. Pests, pathogens and weeds are severe constraints in the productivity and quality of tea. As a result, the tea planters use a wide range of

pesticides to combat these problems for high yield and economic returns. Though broad-spectrum chemicals offer powerful incentives, they have serious drawbacks such as resistance to pesticides, pest resurgence, outbreak of secondary pests, harmful effects on human health and environment due to the presence of undesirable residues. Because of the known toxicity of pesticides in food products (Nagayama et al. 1995; Neidert and Saschenbreker 1996) there is increasing public concern, of late, regarding the pesticide residues in tea also (Singh et al. 1984). In addition, mounting pressure from importing countries as well as environmental health professionals has compelled to assess the impact of pesticide residues in tea as per the recommended maximum residue limits (MRLs). Limited work in this respect describes the determination of organochlorine residues and DDT residues in black tea (Peterson and Jenson 1986). Such information for pesticide residues in Indian tea ecosystems, specifically in the tea growing soils and adjacent water bodies are lacking, although it is known that pesticide residues jeopardize microbial and biochemical aspects of soil quality (Pal et al. 2006).

The present study aimed to investigate the residue level of different organophosphorus (OP), organochlorine (OC), and synthetic pyrethroids (SP) pesticides in made tea, fresh tea leaves, soils and adjacent water bodies of some selected tea gardens in the Dooars (26.44′ 29″ N and 88.38′ 41″ E) and Hill region (26.52′ 48″ N and 88.16′ 48″ E) of West Bengal, India. We studied altogether six tea gardens, which are all conventional tea gardens, in the Dooars. Among the five tea gardens on the Hill regions, that we studied, three were organically managed gardens (as certified by Institute for Marketecology, Switzerland) and the rest were conventionally managed.

Materials and methods

Sampling

Fresh tea leaves and soils were collected from the area consisting of 8–12 year old stands of tea fields from each of the tea garden during the month of April and November 2006, which coincided with the plucking of fresh tea leaves just after pruning in both the seasons. Sampling commenced after 15 to 20 days of pesticide application. Fresh leaves were collected randomly from the upper tier of the tea bushes from six designated sites. For soils, six replicate samples in each area (100 m × 50 m) were taken using a soil corer at the same time. Each replicate consisted of five randomly taken (at the four corners and the center) sub samples of 0–20 cm depth. Six samples of different batches of made tea from the processing units of each of the tea gardens were also collected randomly. Water samples from the free flowing water bodies (wherever found) adjacent to the tea gardens were taken into consideration at different time intervals in a day. All the samples were brought to the laboratory for quantification in airtight sealed containers.

Analytical standards and working solution

The following pesticides commonly used in tea cultivation were selected for the study: organophosphorus (e.g. ethion and chlorpyrifos), organochlorines (e.g. heptachlor, dicofol, α-endosulfan, β-endosulfan and endosulfan sulfate), synthetic pyrethroids (e.g. cypermethrin and deltamethrin). Analytical standards of these pesticides were obtained from ACCU standard, USA (purity >98%). Standard stocks solutions (50 mg/l) were prepared in *n*-hexane. All other solvents and chemicals were of analytical grade from E-Merck (Merck India, Mumbai, India).

Apparatus

A gas–liquid chromatograph Hewlett-Packard 6890 series, equipped with an auto sampler, nitrogen–phosphorus detector (NPD), electron capture detector (ECD) was used. Organophosphorus pesticides were detected with NPD, fused silica semi capillary (HP-5) column (30 m × 0.32 mm × 0.25 μm) was used and the operating conditions were injector temperature 160°C, detector temperature, 300°C, initial oven temperature, 160°C for 1 min, raised at 10°C/min to 250°C, and then held at 250°C for 4 min, again raised to 270°C at 10°C/min and then hold at 270°C for 4 min. The carrier gas was nitrogen at 10 ml/min and the injection volume was 3 μl. Organochlorine and pyrethroids were detected with ECD, fused silica semi capillary (HP-5) column (30 m × 0.32 mm × 0.25 μm) was used and the operating conditions were injector temperature 190°C, detector temperature, 300°C, initial oven

temperature, 190°C for 1 min, raised at 10°C/min to 200°C, and then held at 200°C for 1 min, again raised to 270°C at 10°C/min and then held at 270°C for 5 min. The carrier gas was nitrogen at 10 ml/min and the injection volume was 1 μl.

Detection and quantification

Quantification of pesticides was accomplished using standard curves prepared by diluting the stock solutions in *n*-hexane to levels at which good linearities were achieved in the range of 0.5 to 2.0 ppm with a coefficient of variation between 0.9990 to 0.9997. Overall limit of sensitivity or detection limit for soil, fresh leaves and made tea was 0.01 ppm and for water samples 0.01 ppb respectively.

Sample preparation, extraction and clean up

Pesticide residues in air-dried tea soil (50 g), fresh tea leaves (25 g) and made tea (25 g) were determined by extraction with acetone (200 ml) in Soxlet for 2 h. After centrifugation and concentration with rotary vacuum evaporator at 40°C, the extracts were combined and partitioned with dichloromethane (100+50+50 ml). The aqueous layer was discarded and the organic layer was passed through anhydrous sodium sulfate and the filtrate was recentrifuged and concentrated with rotary vacuum evaporator at 40°C. A chromatographic column was prepacked with anhydrous sodium sulfate, silica gel, florisil (5 g each), and prewashed with *n*-hexane. The sample extract was added to it and the solvent was allowed to settle and run off. The sample extract were eluted off from the column with 200 ml hexane-ethyl acetate (8:2; *v*/*v*) as eluting solvent. The eluted sample was recentrifuged, concentrated to 10 to 20 ml with rotary vacuum evaporator at 40°C and the volume was made up by *n*-hexane (10 ml).

Pesticide residues in water sample (500 ml) were estimated by cleaning up by liquid–liquid partitioning with dichloromethane (100+50+50 ml). The same above process was followed for the extraction of pesticides in water. All the solvents were glass distilled and checked for interference prior to use. Florisil (60–100 mesh), silica gel (60–120 mesh), was activated at 130°C for 24 h and then kept in a dessicator before use. Anhydrous sodium sulfate was heated at 300°C in a furnace for 6 h and stored in a dessicator.

Results and discussion

The positive detections of the pesticides in percent substrate samples collected from the tea gardens in the Dooars and Hill regions are presented in Table 1. The soils, fresh tea leaves and made tea seemed to contain more OP pesticides in higher frequencies than the water bodies in the Dooars region. In contrast the OC pesticides were detected in highest similar frequencies in fresh tea leaves and made tea, followed by soil and water bodies in the Dooars region. The soil samples were devoid of SP pesticides. The fresh tea leaves and made tea samples contained similar frequencies of the SP pesticides in the Dooars. The frequencies of positive detection of the OP pesticides were more or less equal in value in all the substrate samples collected from the tea gardens in the Hill region (Table 1). In case of OC pesticides the highest frequencies were found in soil followed by fresh tea leaves and made tea. The same frequencies of SP were also recorded in the substrate samples of the Hill region as that of the Dooars region.

The estimated amount of the mean total of the pesticide residues in the Dooars region were in descending order of fresh tea leaves (OP: 8.05; OC: 15.08; SP: 0.83 ppm) > soils >(OP: 3.73; OC: 6.68; SP: 0 ppm) > made tea (OP: 2.95; OC: 4.90; SP: 0.34 ppm) > water bodies (OP: 0.0009; OC: 0.0301; SP: 0 ppm; Fig. 1). The lower levels of pesticide residues found in the water bodies than in the soils might be attributed to the fact that the inputs of pesticides to water are a function of suspended particulate concentrations, where the residues were absorbed and transported. These varied from season to season, depending on the rainfall events that control the activities of soil erosion and the amounts of suspended particulates, during runoff (Wan et al. 2005). The estimated amount of mean total of pesticide residues were in the order of fresh tea leaves (OP: 2.38; OC: 3.30; SP: 0.62 ppm) > made tea (OP: 0.61; OC: 1.18; SP: 0.25 ppm) > soils (OP: 0.53; OC: 1.20; SP: 0 ppm) in the Hill region (Fig. 2).

All the samples of made tea, collected from the Dooars region contained residues of both ethion and chlorpyrifos of the OP pesticide group. Residues of

Table 1 Pesticide residues in percentage of substrate samples in the tea gardens of the Dooars and Hill region

Pesticides	Percent samples											
	Made tea				Fresh tea leaves				Soil			Water bodies
	Dooars		Hill		Dooars		Hill		Dooars		Hill	Dooars
	April	November	April	November	April	November	April	November	April	November	April	April	November
Organophosphorus													
Ethion	100	100	40	20	100	100	40	40	100	100	40	33	0
Chlorpyriphos	100	100	40	40	100	100	40	40	100	100	40	0	0
Organochlorine													
Heptachlor	33	100	40	40	33	100	40	40	0	83	40	66	0
Dicofol	33	100	0	40	33	100	0	40	0	100	40	50	0
α-Endosulfan	83	50	40	40	83	50	40	40	83	33	40	16	0
β-Endosulfan	100	100	40	40	100	100	40	40	100	83	40	16	0
Endosulfan sulfate	66	66	40	40	66	66	40	40	66	66	40	0	0
Pyrethroid													
Cypermethrin	0	100	40	40	0	100	40	40	0	0	0	0	0
Deltamethrin	0	83	20	40	0	83	20	40	0	0	0	0	0

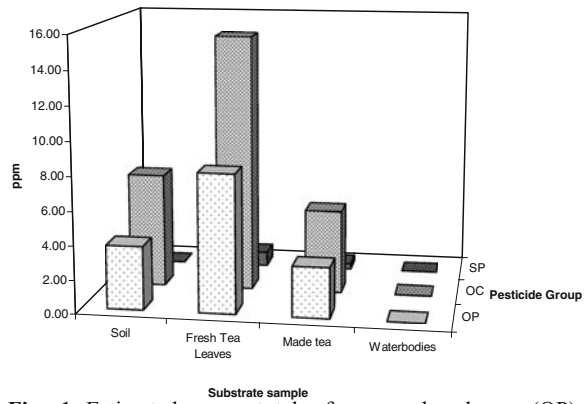

Fig. 1 Estimated mean total of organophosphorus (OP), organochlorine (OC) and synthetic pyrethroid (SP) pesticides in the Dooars region

ethion were higher in April than that of chlorpyrifos. The concentrations of ethion in made tea samples were within specified MRL (European Union 2007), but chlorpyrifos residue concentrations marginally exceeded the MRL in April (Table 4). The estimated mean total of OP pesticide concentrations was higher in April than in November. The concentrations of ethion and chlorpyrifos residues in fresh tea leaves followed the same trend as in made tea. However, the concentrations of OP pesticide residues in made tea were lower compared to fresh tea leaves, which corroborated the previous findings of (Muraleedharan 1994). Fresh tea leaves after plucking generally are subjected to a manufacturing process that involves steps such as withering, rolling and drying. There are ample chances for loss of pesticides in all the steps involved in conventional manufacture (Sood et al.

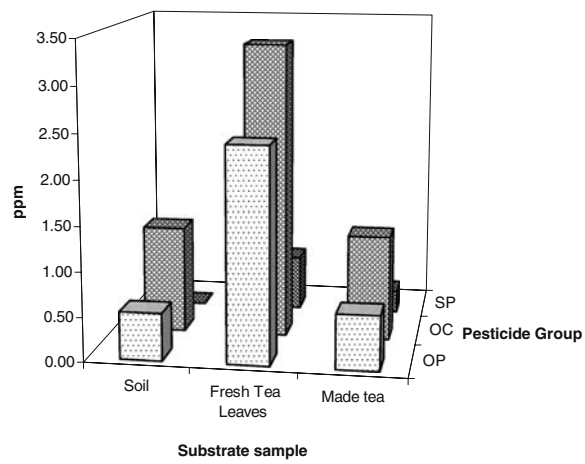

Fig. 2 Estimated mean total of organophosphorus (OP), organochlorine (OC) and synthetic pyrethroid (SP) pesticides in the Hill region

Table 2 Organophosphorus (OP), organochlorine (OC) and synthetic pyrethroid (SP) pesticide residues (means, ranges in parentheses) in various substrate samples of selected tea gardens in the Dooars region

Pesticides	Made tea (dry wt.), ppm		Fresh tea leaves (dry wt.), ppm		Soil (dry wt.), ppm		Water bodies, ppm	
	April	November	April	November	April	November	April	November
Organophosphorus								
Ethion	0.247 (0.054–1.0701) 36/36[b]	0.0648 (0.016–0.2562) 36/36	0.671 (0.0188–2.82) 36/36	0.248 (0.0107–0.9999) 36/36	0.161 (0.0109–0.5308) 36/36	0.210 (0.0131–0.899) 36/36	0.0005 (0.0003–0.0005) 12/36	BDL[a]
Chlorpyrifos	0.102 (0.0246–0.4328) 36/36	0.0728 (0.0249–0.1151) 36/36	0.194 (0.0554–0.7299) 36/36	0.228 (0.1036–0.4563) 36/36	0.110 (0.0166–0.5301) 36/36	0.141 (0.0739–0.2731) 36/36	BDL	BDL
Estimated mean total	0.349	0.137	0.865	0.476	0.271	0.351	0.0005	
Organochlorine								
Heptachlor	0.0709 (0.0332–0.1095) 12/36	0.203 (0.0719–0.5251) 36/36	0.923 (0.3245–1.5234) 12/36	0.540 (0.2357–0.8288) 36/36	BDL	0.238 (0.1638–0.4259) 30/36	0.0043 (0.0008–0.0076) 24/36	BDL
Dicofol	0.0327 (0.0272–0.0384) 12/36	0.219 (0.0933–0.6525) 36/36	0.078 (0.0602–0.0953) 12/36	0.517 (0.2622–0.9431) 36/36	BDL	0.450 (0.221–0.896) 36/36	0.0036 (0.0009–0.0058) 18/36	BDL
α-Endosulfan	0.0471 (0.0278–0.0912) 30/36	0.0628 (0.0249–0.1109) 18/36	0.176 (0.0962–0.2248) 30/36	0.155 (0.0144–0.3218) 18/36	0.0549 (0.025–0.0799) 30/36	0.1466 (0.0933–0.1999) 12/36	0.0005 (0.0004–0.0005) 6/36	BDL
β-Endosulfan	0.170 (0.0431–0.2858) 36/36	0.0696 (0.0127–0.2019) 36/36	0.344 (0.0917–0.5219) 36/36	0.360 (0.0139–0.9238) 36/36	0.092 (0.074–0.1574) 36/36	0.227 (0.0112–0.7601) 30/36	0.0016 (0.0011–0.0022) 6/36	BDL
Endosulfan	0.0405 (0.0276–0.0696)	0.0330 (0.0148–0.066)	0.154 (0.1224–0.2358)	0.137 (0.0733–0.229)	0.0790 (0.048–0.0969)	0.0471 (0.0188–0.0977)	BDL	BDL
Sulfate	24/36	24/36	24/36	24/36	24/36	24/36		
Estimated mean total	0.361	0.587	1.521	1.709	0.226	1.112	0.01	
Pyrethroid								
Cypermethrin	BDL	0.033 (0.0187–0.0456) 36/36	BDL	0.091 (0.0.0131–0.2546) 36/36	BDL	BDL	BDL	BDL
Deltamethrin	BDL	0.0299 (0.0205–0.0374) 30/36	BDL	0.0571 (0.0372–0.0656) 30/36	BDL	BDL	BDL	BDL
Estimated mean total		0.0629		0.1481				

[a] Below detection limit

[b] $n/36$; where n=number of samples above detection limit

Table 3 Organophosphorus (OP), organochlorine (OC) and synthetic pyrethroid (SP) pesticide residues (means, ranges in parentheses) in various substrate samples of selected tea gardens in the Hill region

Pesticides	Made tea (dry wt.), ppm		Fresh tea leaves (dry wt.), ppm		Soil (dry wt.), ppm	
	April	November	April	November	April	November
Organophosphorus						
Ethion	0.0181 (0.015–0.0267) 12/30[a]	0.0852 (0.0844–0.859) 6/30	0.0407 (0.0217–0.0594) 12/30	0.0683 (0.0251–0.1128) 12/30	0.025 (0.018–0.0329) 12/30	0.043 (0.0178–0.062) 12/30
Chlorpyrifos	0.1368 (0.0515–0.2219) 12/30	0.109 (0.0741–0.1431) 12/30	0.600 (0.1092–1.0902) 12/30	0.481 (0.1652–0.7952) 12/30	0.019 (0.0146–0.0256) 12/30	0.177 (0.0952–0.2607) 12/30
Estimated mean total	0.155	0.194	0.64	0.549	0.044	0.22
Organochlorine						
Heptachlor	0.0388 (0.0295–0.0478) 12/30	0.0195 (0.0151–0.0235) 12/30	0.11 (0.0943–0.1253) 12/30	0.30 (0.2088–0.3917) 12/30	0.0805 (0.0775–0.0826) 12/30	0.155 (0.1487–0.1613) 12/30
Dicofol	BDL[b]	0.2506 (0.2475–0.2537) 12/30	BDL	0.5144 (0.5117–0.5133) 12/30	BDL	0.187 (0.0641–0.3112) 12/30
α-Endosulfan	0.0196 (0.0151–0.0245) 12/30	0.1548 (0.1411–0.1687) 12/30	0.0484 (0.0417–0.0557) 12/30	0.387 (0.3611–0.4130) 12/30	0.025 (0.0195–0.0312) 12/30	0.035 (0.0295–0.0399) 12/30
β-Endosulfan	0.0149 (0.0105–0.0199) 12/30	0.124 (0.094–0.0223) 12/30	0.0560 (0.0530–0.0592) 12/30	0.0718 (0.0442–0.0996) 12/30	0.011 (0.0108–0.0142) 12/30	0.035 (0.0311–0.0398) 12/30
Endosulfan sulfate	0.0316 (0.0226–0.0419) 12/30	0.0518 (0.0392–0.0859) 12/30	0.0572 (0.0415–0.0728) 12/30	0.1059 (0.0106–0.2011) 12/30	0.029 (0.0262–0.0326) 12/30	0.040 (0.027–0.0661) 12/30
Estimated mean total	0.105	0.489	0.271	1.379	0.147	0.452
Pyrethroid						
Cypermethrin	0.0165 (0.0132–0.0197) 12/30	0.0392 (0.0114–0.0676) 12/30	0.0762 (0.0736–0.0798) 12/30	0.0811 (0.0388–0.1234) 12/30	BDL	BDL
Deltamethrin	0.0148 (0.0104–0.0204) 12/30	0.0579 (0.0565–0.0593) 12/30	0.0741 (0.0733–0.0745) 6/30	0.1161 (0.1117–0.1207) 12/30	BDL	BDL
Estimated mean total	0.0313	0.0971	0.15	0.197		

[a] *n*/30; where *n*=number of samples above detection limit

[b] Below detection limit

Table 4 Percentage of made tea samples above MRL level of pesticide residues

Pesticides	Dooars		Hill	
	April	November	April	November
Organophosphorus				
Ethion	0	0	0	0
Chlorpyriphos	16	16	20	20
Organochlorine				
Heptachlor	33	100	40	20
Dicofol	0	0	0	0
α-Endosulfan	0	0	0	0
β-Endosulfan	0	0	0	0
Endosulfan sulfate	0	0	0	0
Pyrethroid				
Cypermethrin	0	0	0	0
Deltamethrin	0	0	0	0

2004). The concentrations of these two pesticide residues in soils were lower in comparison to fresh tea leaves. Unlike tea samples, estimated amount of mean total of OP pesticide residues in soil recorded higher value in November than April. Ethion was detected in some of the water bodies in April only (Table 2). None of the OC pesticide residues were detected in water bodies in November samples (Table 2). The estimated amount of mean total of OC pesticide residues in made tea, soil samples and fresh tea leaves were higher in November compared to April. Among the OC pesticide residues, dicofol recorded the highest value in November samples of made tea. Excepting heptachlor, none of the studied OC pesticide residues crossed the MRL (Table 4). In this case also, concentrations of OC pesticide residues in fresh tea leaves were very much higher compared to made tea.

Among the residues of SP, cypermethrin and deltamethrin could not be detected in made tea and fresh tea leaves in April. However the estimated amounts of mean total of these two pyrethroids were higher in fresh tea leaves than made tea in November. These two pesticides were also not detected in soils as well as in the water bodies in both April and November. The estimated mean total of OP pesticide residues in made tea and soil but not in fresh tea leaves was higher in November compared to April in the Hill region (Table 3). The levels of the two OP pesticides in made tea were much lower compared to fresh tea leaves in both the seasons. The residue concentrations in soil were much lower with respect to tea samples. The estimated mean total of OC pesticide residues was much higher in November than April in all the samples. Dicofol could not be detected in the samples of made tea, fresh tea leaves and soils in April. The SP pesticide residues were not detected in soil during both the sampling periods. However, the estimated mean total of the SP group was higher in fresh tea leaves than made tea in both the seasons (Table 3).

The Table 4 represents the percentage of made tea samples above MRL level of pesticide residues (European Union 2007). Sixteen percent of the made tea samples exceeded the MRL level of chlorpyrifos both during April and November. The residues of heptachlor exceeded in 33% and 100% samples in April and November respectively in the Dooars region. Forty percent of the made tea samples in April and 20% samples in November exceeded the MRL value of Heptachlor in the Hill region. Chlorpyrifos exceeded the MRL level in 20% of the samples collected from the Hill region.

Conclusions

The present study reveals that some of the tea gardens in West Bengal receive indiscriminate input of pesticides, to boost its production at the cost of undesirable residues in the environment. It is indicated in the study that heptachlor and chlorpyrifos pesticides despite being banned, are prevalent in made tea at

higher concentrations than their respective MRLs. This may pose serious health hazards to the consumers apart from losing export credentialities.

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