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Determination of Organochlorine and Synthetic Pyrethroid Pesticide Residues in Water Samples Collected from Different Locations of Bangladesh

Mohammad Dalower Hossain Prodhan*, Md. Sultan Ahmed, Nirmal Kumar Dutta, Debasish Sarker, Syed Nurul Alam

Pesticide Analytical Laboratory, Pesticide Research and Environmental Toxicology Section, Entomology Division, Bangladesh Agricultural Research Institute, Gazipur, Bangladesh
Email: *dalowerag@yahoo.com, *mdhprodhan@gmail.com

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Abstract

The present study was aimed to validate an analytical method for the quantification of 19 organochlorine and 2 synthetic pyrethroid pesticide residues in water samples using modified quick, easy, cheap, effective, rugged and safe (QuEChERS) extraction and Gas Chromatography coupled with Electron Capture Detector (ECD). The selected pesticide residues were determined by in-house validated method. The analytical method was validated by evaluating the accuracy, precision, linearity, limit of detection (LOD) and limit of quantification (LOQ). The average recoveries of the selected pesticides ranged from 78% to 117% with RSDr ≤ 12% in two fortification levels of 0.02 and 0.1 mg/L. The linearity was ≥0.995 for all of the selected pesticides. The LOD ranged from 0.003 to 0.006 mg/L and the LOQ was 0.02 mg/L for all the selected analytes. This method was applied satisfactorily for the residue analysis of 108 water samples collected from nine districts of Bangladesh. Among the analyzed samples, only 4 had cypermethrin residues (0.026 mg/L, 0.034 mg/L, 0.045 mg/L and 0.05 mg/L). The level of detected cypermethrin residues were above the WHO recommended guide line values of water quality.

Keywords

Organochlorine Pesticides, Synthetic Pyrethroid Pesticides, Water Samples, Residue Analysis, Gas Chromatography

1. Introduction

Water is one of the most important substances on earth. All plants and animals

must have water to survive. If there was no water, there would be no life on earth. However, waters are contaminated by pesticides through percolation and runoff from agricultural land and channels, and from urban city sewage sites, thus affecting the quality of various water sources. The organochlorine pesticide (OCP) residues are still detectable in water due to their long persistence in the environment. Several works have indicated the presence of OCPs residues in waters [1] [2] [3].

Up until now, in Bangladesh, the farmers mostly depend on toxic chemical pesticides to prevent the crop loss caused by insect pests infestation and to increase the production. There are many reasons behind this. Pesticide adulteration is one of the major reasons for the indiscriminate use of pesticides. Due to adulteration, the activity of pesticides is reduced and that is why, the farmers are applying pesticides too often for the management of insect-pests and diseases as well as to control the weeds. As a result of frequent application of pesticides, their residues are remaining of different agricultural commodities.

Till today, in Bangladesh several researchers have been detected pesticide residues in vegetables [4]-[16], fruits [17], betel leaf [18], sugarcane [19], dry fish [20] and fish [21] [22]. Therefore, it is inevitable that the pond water, fresh water as well as the ground water may be contaminated with the residues of such toxic chemical pesticides used in the above mentioned crops by the farmers of Bangladesh.

In the analysis of pesticide residues, effective extraction and clean-up methods are essential. Nowadays, the quick, easy, cheap, effective, rugged and safe (QuECHERS) method is widely used for extraction and cleanup of pesticide residues in a wide variety of matrices [23] [24] [25] [26]. This method is gaining popularity day by day compared to the other existing methods as it has a lot of advantages. The important ones are high recoveries of analytes with low organic solvent consumption and the low cost per sample. Gas Chromatography coupled with Electron Capture Detector (GC-ECD) is widely used for the quantification of organochlorine and synthetic pyrethroid pesticides as they are very sensitive for the above mentioned groups of pesticides. On the other hand, GC-ECD are cheaper and have lower maintenance costs, hence are more readily available in some countries than mass spectrometers [17]. Therefore, the QuECHERS extraction technique followed by Gas Chromatography is a valuable tool for the quantification of organochlorine and synthetic pyrethroid pesticide residues in water samples.

The organochlorine pesticides create several adverse effects on human heath, and their associated health risk are monitored by several researchers [27] [28]. Keeping this view the present study was initiated to develop and validate an analytical method for the quantification of 19 organochlorine pesticides (alpha BHC, delta BHC, beta BHC, gama BHC, heptachlor, aldrin, heptachlor epoxide, gama chlordane, alpha chlordane, alpha endosulfan, 4,4 DDE, dieldrin, endrin, 4,4 DDD, beta endosulfan, 4,4 DDT, endrin aldehyde, endosulfan sulphate, methoxychlor, and endrin ketone) and 2 synthetic pyrethroid pesticides residues (cy-

permethrin, fenvalerate) and to monitor the selected organochlorine and synthetic pyrethroid pesticide residues in water samples collected from Bogura, Cumilla, Dhaka, Gazipur, Jamalpur, Jashore, Mymensingh, Narsingdi and Rangpur district of Bangladesh.

2. Materials and Methods

2.1. Chemicals & Reagents

Reference standards of 19 organochlorine pesicide Mix (alpha BHC, delta BHC, beta BHC, gama BHC, heptachlor, aldrin, heptachlor epoxide, gama chlordane, alpha chlordane, alpha endosulfan, 4,4 DDE, dieldrin, endrin, 4,4 DDD, beta endosulfan, 4,4 DDT, endrin aldehyde, endosulfan sulphate, methoxychlor, and endrin ketone) and 2 synthetic pyrethroid pesticide (cypermethrin, fenvalerate) were obtained from SIGMA-Aldrich, Germany through SF Scientific, Dhaka. Analytical grade acetonitrile (MeCN), methanol, sodium chloride (NaCl), anhydrous magnesium sulphate ($MgSO_4$) and Primary Secondary Amine (PSA) were obtained from SIGMA-Aldrich, Germany through SF Scientific, Dhaka.

2.2. Preparation of Pesticide Standard Solution

Mixed pesticide standard stock solutions (200 ug/mL) of alpha BHC, delta BHC, beta BHC, gama BHC, heptachlor, aldrin, heptachlor epoxide, gama chlordane, alpha chlordane, alpha endosulfan, 4,4 DDE, dieldrin, endrin, 4,4 DDD, beta endosulfan, 4,4 DDT, endrin aldehyde, endosulfan sulphate, methoxychlor, and endrin ketone were prepared in hexane:toluene (50:50) except for cypermethrin and fenvalerate. In case of cypermethrin and fenvalerate it was acetonitrile. An intermediate mixed standard solution of 10 ug/mL in acetonitrile was prepared from the mixed standard solution of 200 ug/mL. Then working standard solutions of 0.1, 0.2, 0.5, 1.0, 2.0, and 3.0 ug/mL in acetonitrile were prepared by transferring the appropriate amount from 10 ug/mL intermediate mixed standard solution into six separate 5-mL volumetric flasks. The pesticide standard solutions were prepared in the normal room temperature, after the preparation of standard solutions, all the standard solutions were kept in a freezer at −20˚C until use.

2.3. Sample Preparation

The quick, easy, cheap, effective, rugged and safe (QuEChERS) extraction technique, which was first introduced by Anastassiades and their associates [29], is widely used for the extraction and cleanup of pesticide residues for a wide variety of matrices. The QuEChERS extraction method modified by Prodhan and their associates [30], was used for the extraction and clean-up of the selected pesticides from water samples. In brief, 10 mL of water sample was taken in a 50 mL screw-capped polypropylene centrifuge tube and 10 mL acetonitrile (MeCN) was added into the centrifuge tube. The centrifuge tube was closed properly and shaken vigorously for 30 sec. by vortex mixer. Then 4 g anhydrous $MgSO_4$, 1 g

NaCl were added into the centrifuge tube and it was shaken by vortex mixer for 1 minute. Afterwards, the extract was centrifuged for 5 min at 5000 rpm. An aliquot of 3 mL of the MeCN layer was transferred into a 15 mL micro centrifuge tube containing 600 mg anhydrous $MgSO_4$ and 120 mg Primary Secondary Amine (PSA). The content of the centrifuge tube was thoroughly mixed by vortex for 30 sec. and centrifuged for 5 minutes at 4000 rpm. After centrifuge, a 1 mL supernatant was filtered by a 0.2 μm PTFE filter, and then it was taken in a clean GC vial for injection.

2.4. Operating Condition of GC

The concentrations of the extracted organochlorine and synthetic pyrethroid pesticides was detected by GC-ECD (Shimadzu GC-2010), coupled with a capillary column of 30 m long × 0.32 mm ID × 0.25 μm film thicknesses (Rtx-CLPesticides2). The GC-ECD was handled in split mode. Nitrogen was used as carrier (column flow rate 2.71 $mL \cdot min^{-1}$) and make up gas (flow rate-15 $mL \cdot min^{-1}$).

In case of organochlorine pesticides, the injector and detector temperatures were set to 250˚C and 330˚C, respectively and the column oven temperature was programmed, which was started from 180˚C and went up to 220˚C with an incremental rate of 5˚C (12 min hold), then it raised to 260˚C with an incremental rate of 5˚C. All the injections (1 μL) were done in spit mode. The total run time was 28 min.

However, in case of cypermethrin, the injector and detector temperatures were set to 280˚C and 300˚C, respectively, and the column oven temperature was programmed as follows: initial temperature of 160˚C was held for 1 min and ramped to 270˚C with an incremental rate of 10˚C was held for 8 min, resulting the total run time of 20 min. While, in case of fenvalerate, the injector and detector temperatures were set to 280˚C and 300˚C, respectively, and the column oven temperature was programmed as follows: initial temperature of 160˚C and ramped to 230˚C with an incremental rate of 10˚C, and finally went to 270˚C with an incremental rate of 2˚C resulting the total run time of 27 min. Identification of the analyte in the extracted samples was done by comparing the retention time of the corresponding calibration standard and quantification was done by the external calibration curves maid with 6 point calibration standard.

3. Results and Discussions

3.1. Method Validation

The analytical method was validated by evaluating the accuracy, precision, limit of detection, limit of quantification and linearity.

3.1.1. Accuracy and Precision

The accuracy of the method was calculated as % recovery of pesticides from spiked samples. Pesticide free 10 mL water sample was spiked prior to the determination procedure by the addition of a mixed pesticide standard working solution to reach the final fortification levels of 0.02, and 0.10 mg/L. For each

level, five replicates were analyzed. After the addition of each concentration in the matrix, the mixture was equilibrated by shaking and the samples were allowed to settle for 30 minutes prior to extraction in order to ensure the sufficient contact of the analytes with the whole matrix. Then, the samples were prepared according to the method which was described earlier. Precision in case of repeatability (RSD_r) was determined at two fortification levels of 0.02, and 0.10 mg/L with 5 replicates on the same day. A very good accuracy and precision was found for all of the analytes. The average recoveries ranged from 78% to 117% with relative standard deviations (RSD_r) ≤ 12% for all of the analytes (Table 1).

3.1.2. Limit of Detection (LOD) and Limit of Quantification (LOQ)

The LOD was calculated according to EURACHEM guidelines [31]. In order to determine the LOD of each analyte 10 independent sample blanks fortified at the lowest acceptable concentration of 0.02 mg/L were injected and the LOD was expressed as the analyte concentration corresponding to 3 times the standard

Table 1. Mean recovery (%) and RSD (%) of the selected pesticides in water samples.

Pesticides	Fortification level			
	0.02 mg/L		0.1 mg/L	
	Mean (%)	RSD (%)	Mean (%)	RSD (%)
Alpha BHC	82	4	87	6
Delta BHC	85	7	81	9
Beta BHC	84	3	93	4
Gama BHC	117	5	116	7
Heptachlor	86	8	88	5
Aldrin	78	7	79	5
Heptachlor Epoxide	82	5	82	5
Gama Chlordane	80	6	87	9
Alpha Chlordane	81	3	89	8
Alpha Endosulfan	82	9	102	8
4,4 DDE	83	8	84	6
Dieldrin	84	11	89	9
Endrin	86	7	89	12
4,4 DDD	88	9	90	9
Beta Endosulfan	86	6	93	7
4,4 DDT	86	5	87	4
Endosulfan sulphate	113	4	110	8
Methoxychlor	86	8	88	7
Endrin ketone	85	10	81	6
Cypermethrin	97	6	105	6
Fenvalerate	102	5	94	8

deviation. The LOQ was determined according to the European Commission (EC) document no. SANTE/12682/2019 [32]. The LOQ was set at the lowest fortification level for each pesticides that was achieved the acceptable accuracy (mean recoveries for individual pesticides in the range of 70% - 120%) and precision (RSDr ≤ 20%).

The LOD of each analyte is presented in Table 2. The LOD ranged from 0.003 to 0.006 mg/L. The LOQ for all of the selected pesticides was set to 0.02 mg/L which was achieved the acceptable accuracy (mean recoveries for individual pesticides in the range of 78% to 117%) and precision (RSDr ≤ 11%).

3.1.3. Calibration Curve & Linearity

Six point calibration curves were prepared by working standard solutions of 0.01, 0.02, 0.05, 0.10, 0.20, and 0.30 ug/mL in acetonitrile and analyzed in triplicate. Calibration curves were made by plotting the mean peak area of the selected pesticides versus concentration. Linearity was very good and coefficients of determination were ≥0.995 for all of the selected pesticides. The correlation

Table 2. Retention time (RT), limit of detection (LOD), limit of quantification (LOQ) and coefficient of determination (R^2) of the selected pesticides in water.

Pesticides		RT	LOD (mg/L)	LOQ (mg/L)	R^2
Organochlorine	Alpha BHC	5.48	0.003	0.02	0.995
	Delta BHC	6.33	0.003		0.997
	Beta BHC	6.55	0.005		0.995
	Gama BHC	7.33	0.004		0.996
	Heptachlor	7.46	0.005		0.998
	Aldrin	8.36	0.004		0.997
	Heptachlor Epoxide	10.31	0.005		0.998
	Gama Chlordane	11.07	0.004		0.999
	Alpha Chlordane	11.73	0.005		0.996
	Alpha Endosulfan	11.97	0.005		0.998
	4,4 DDE	12.67	0.004		0.996
	Dieldrin	13.37	0.005		0.996
	Endrin	15.15	0.006		0.997
	4,4 DDD	16.33	0.006		0.997
	Beta Endosulfan	16.69	0.005		0.998
	4,4 DDT	18.83	0.006		0.996
	Endosulfan sulphate	22.05	0.006		0.998
	Methoxychlor	24.96	0.005		0.995
	Endrin ketone	25.67	0.006		0.997
Synthetic Pyrethroid	Cypermethrin	12.81	0.005	0.02	0.996
	Fenvalerate	24.46	0.006		0.997

coefficients for all of the selected pesticides are summarized in Table 2.

3.2. Application of the Method for Real Sample Analysis

The concentrated extracts of water samples collected from different locations (Bogura, Cumilla, Dhaka, Gazipur, Jamalpur, Jashore, Mymensingh, Narsingdi and Rangpur) were analyzed by GC-2010 (Shimadzu) with Electron Capture Detector (ECD) with the pre-set parameters. The level of detected pesticide residues found in the analyzed samples is outlined in Table 3. A total of 108 samples were collected from 9 different districts of Bangladesh and were analyzed. Out of 108 analyzed samples, only 4 (3.7% of the analyzed samples) had cypermethrin residues. The level of cypermethrin residues were 0.026 mg/L, 0.034 mg/L, 0.045 mg/L and 0.05 mg/L, respectively. The level of detected cypermethrin residues were above the WHO recommended guide line values of water quality. The results of the present study are in a good agreement with the findings of Uddin and their associates [33]. They have also found that cypermethrin was the most frequently found pesticide in the analyzed water samples. It is a good indication for the peoples of Bangladesh that none of the analyzed samples were found contaminated with toxic organochlorine pesticides. On the other hand, only 3.7% analyzed samples were found contaminated with the residues of cypermethrin.

The water samples collected from Bogura, Cumilla, Dhaka, Jamalpur, Jashore, Narsingdi and Rangpur did not contained any residues of the 21 analyzed pesticides. The four contaminated samples were collected from Gazipur and Mymensingh. Among these 4 contaminated samples, 3 were collected from Mymensingh and 1 was from Gazipur. The samples collected from Bogura, Cumilla, Dhaka, Jamalpur, Jashore, Narsingdi and Rangpur did not contained any residues might

Table 3. Residue levels (mg/L) found in water samples collected from different locations of Bangladesh.

Area of collection	Analyzed samples (No.)	Contaminated samples (No.)	Detected pesticides	Residue levels (mg/L)	WHO recommended value (mg/L)
Bogura	12	-	ND*	-	-
Cumilla	12	-	ND*	-	-
Dhaka	12	-	ND*	-	-
Gazipur	12	1	Cypermethrin	0.05	0.02
Jamalpur	12	-	ND*	-	-
Jashore	12	-	ND*	-	-
Mymensingh	12	3	Cypermethrin	0.026-0.045	0.02
Narsingdi	12	-	ND*	-	-
Rangpur	12	-	ND*	-	-
Total	108	4			

ND*—Not Detected.

be the reason of sample type as from these locations, only the drinking water was collected. However, in case of Mymensingh and Gazipur along with the drinking water, pond water samples were also collected, and from these both locations, the contaminated samples were originated from the pond water samples. Therefore, it is necessary to collect and analyze the pond water samples from different locations of the country as the farmers of Bangladesh are using different pesticides for the production of rice and vegetables. Hence, the residues of pesticides may remain in pond water adjacent to the rice and vegetable field. In our future study, emphasis should be given to analyze pond and river water as well as fresh water samples in order to find out the actual scenario of water contamination by different pesticides.

4. Conclusion

The proposed method in this study is an efficient, easy and effective multi-residue analytical method for the analysis of 19 organochlorine and 2 synthetic pyrethroid pesticide residues in water samples using Gas Chromatography coupled to Electron Capture Detector (GC-ECD). In this study, a very good accuracy and precision was found for all the analytes. The average recoveries ranged from 78% to 117% with $RSD_r \leq 12\%$ in two fortification levels of 0.02 and 0.1 mg/L. The linearity was ≥0.995 for all of the selected pesticides. The LOD ranged from 0.003 to 0.006 mg/L and the LOQ was 0.02 mg/L. Moreover, a total of 108 water samples collected from nine districts of Bangladesh were analyzed successfully using this proposed analytical method. Among the analyzed samples, only 4 had cypermethrin residues (0.026 mg/L, 0.034 mg/L, 0.045 mg/L and 0.05 mg/L). The level of detected cypermethrin residues were above the WHO recommended guide line values of water quality [34]. Thus, the proposed method can be used successfully to monitor organochlorine and synthetic pyrethroid pesticide residues in water samples.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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