RESEARCH ARTICLE

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Assessment of Pesticide Residues in Organic Rice Production in The Mekong Delta, Vietnam

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ABSTRACT

Organic rice production has been developed in Mekong Delta to reduce toxicity risk for humans and the environment, however, some models have failed due to pesticide residues that were not investigated in advance. So, it is important to test for prohibited residual pesticides in organic farming. The rice samples in ten districts of six provinces in Mekong Delta, Vietnam, were sampled and analyzed to check the presence of 854 substrates that are prohibited for use in organic production. A total of 18 active ingredients were detected. Based on the contamination rates, they are divided into three groups. The high presence group includes only tricyclazole (62.5%). The medium presence group includes difenoconazole (32.5%), chlorpyrifos-ethyl (30%), isoprothiolane (25%), propiconazole (20%), azoxystrobin (17.5%), hexaconazole (15%), chlorfenapyr (15%), cypermethrin (10%). The low presence group includes paclobutrazol (7.5%), tebuconazole (5%) and the seven remaining ingredients niclosamide, chlorfenson, fipronil, fipronil-desulfinyl, fenoxanil, metalaxyl & metalaxyl-M, and chlorfluazuron with the same contamination rate (2.5%). Based on the present period, they are divided into four groups. The four-year presence group includes tricyclazole, difenoconazole, and propiconazole. The three-year presence group includes chlorpyrifos-ethyl, isoprothiolane, hexaconazole, chlorfenapyr, and cypermethrin. The two-year presence group includes azoxystrobin and paclobutrazol. The one-year presence group includes the eight active ingredients with the lowest contamination rate as tebuconazole, niclosamide, chlorfenson, fipronil, fipronil-desulfinyl, fenoxanil, metalaxyl & metalaxyl-M, and chlorfluazuron. Among 18 detected active ingredients, there were 8 substances used for disease control, 8 substances used for insecticides, 1 substance used for killing snails, and 1 substance used for growth regulation. This finding is useful for developing organic rice zones in the Mekong Delta.

Keywords: Mekong Delta, Organic Rice Farming, Pesticide Residues, Tricyclazole

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I. INTRODUCTION

Rice production in the Mekong Delta plays an important role in food production and the country's broader economy. The agricultural land accounts for 64% of the total area (GSO, 2018), with rice being the most important crop, accounting for 73.9% of the agricultural land (Nguyen *et al.*, 2015). The Mekong Delta is a "rice bowl" of Vietnam, accounting for around 57% of national rice production and 90% of total export volume (GSO, 2015; Tong, 2017). The domestic and export market for rice from the Mekong Delta contributes a fifth to the GDP of the country (Nguyen *et al.*, 2015). However, to maintain national food security and Vietnam's leading position as one of the largest rice exporters worldwide, paddy rice production has intensified over the last decades (Tong, 2017). This intensification increased the adoption of high yield varieties and increased fertilizer and pesticide applications (Phong *et al.*, 2010; Anyusheva *et al.*, 2012), causing widespread pollution in surface water bodies, drinking water resources, soil and sediments (Toan *et al.*, 2013; Chau *et al.*, 2015).

The increased reliance on pesticides in rice production has, in some areas, proved to be unsustainable and cost-ineffective due to pesticide-induced outbreaks of insect pests, the development of pesticide-resistant pests, the rising cost of pesticide use, and the negative effects of pesticide use on human health and the environment (Le & Pham, 2012; Pimentel & Burgess, 2014; Mnif *et al.*, 2011; Zheng *et al.*, 2016). That is the reason why pesticide residues in food are a major public health concern that harms both producers and consumers (Hou *et al.*, 2013).

Since 2015, several organic rice production models have been built in the Mekong Delta to help farmers apply new farming techniques, replacing harmful chemical inputs with biological and organic derived substances (Nguyen & Van, 2021). Organic farming improves the health of soil organisms, plants, livestock, and people within the agro-ecosystem and helps build environmentally-friendly businesses (Martin, 2009). However, the biggest challenge in organic agriculture is changing farmers' mindsets and farming practices that favour the application of toxic chemicals. In some cases, the farmers did not follow organic production procedures to use the prohibited pesticides on their farms. This has led to the failure of the organic rice models, affecting the entire project and causing financial damage to the invested enterprises (Nguyen & Van, 2021). So, evaluating the presence of prohibited residual pesticides in organic production is necessary to develop organic rice production in Mekong Delta.

The aims of the study were (i) to investigate pesticide residues used in organic production (ii) to document the common pesticide residues in organic rice production both in rice-intensive farming and rice-shrimp integrated farming systems in the Mekong Delta.

II. MATERIALS AND METHODS

A. *Study Sites*

The Mekong Delta is the largest agricultural region in Vietnam, covering 39,000 km2 (Schmidt, 2020). The Mekong Delta lies within the humid tropics, characterized by consistently high mean monthly temperatures (25 –29^{o}C). The Asian monsoons predominantly control seasonal climatic variations. The average annual rainfall is approximately 1800 mm, and the monthly temperature is 25 –29^{o}C, with a wet season from May to November (Hashimoto & Takehiko, 2011; MDP, 2011). For the purposes of this study, study sites were located in ten districts of six provinces where rice is cultivated in both rice-intensive farming and rice-shrimp integrated farming systems. In permanent rice areas, including Tri Ton and Thoai Son districts of An Giang province, Tan Hiep district of Kien Giang province, Nga Nam town of Soc Trang province, and Vung Liem district of Vinh Long province, paddy rice is cultivated with two or three harvests per year. In the alternating rice-shrimp systems, including Cau Ngang district of Tra Vinh province, Thoi Binh district of Ca Mau province, U Minh Thuong district of Kien Giang province, My Xuyen and Thanh Tri districts of Soc Trang province, shrimp and rice production are alternated between the dry and the wet season, respectively (Table I and Fig. 1).

TABLE I: SAMPLING SITES AND TIME

No.	Sampling sites	Abbreviation	Sampling time	No. of samples
1	Cau Ngang district, Tra Vinh provinces	CN-TV	14/02/2017	3
			26/02/2017	1
			10/04/2018	1
2	Thoi Binh district, Ca Mau province	TB-CM	19/12/2018	2
			29/12/2018	1
3	Tri Ton district, An Giang province	TT-AG	07/03/2019	3
			04/09/2019	1
			23/12/2019	2
			20/03/2020	1
			31/03/2020	1
			27/11/2020	2
			17/08/2021	1
			24/08/2021	2
			14/09/2021	1
4	Thoại Son district, An Giang province	TS-AG	14/04/2021	1
5	Tan Hiep district, Kien Giang province	TH-KG	01/08/2019	1
6	U Minh Thuong district, Kien Giang province	UMT-KG	19/09/2019	1
			27/01/2021	4
7	Nga Nam town, Soc Trang province	NN-ST	22/05/2019	1
			27/05/2019	1
8	My Xuyen district, Soc Trang province	MX-ST	01/06/2019	3
9	Thanh Tri district, Soc Trang province	TT-ST	25/12/2020	2
10	Vung Liem district, Vinh Long province	VL-VL	22/05/2019	1
			04/02/2021	1
			27/05/2021	2

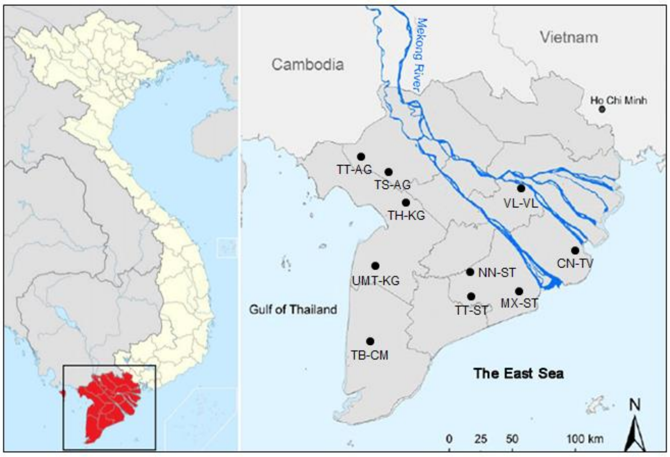

Fig. 1. Location of the study sites.

B. Studied Pesticides and Rice Cultivation

The study pesticides were selected according to the audit program of the organic certification body, ControlUnion (CU). According to the audit program of CU, the total number of investigated pesticides was 854 substrates, as reported in our previous study (Duong *et al.*, 2022). Rice varieties grown in these studied areas were Hong Ngoc Oc Eo and ST25, DS1, ST24, and Dai Thom 8, which grew around 95 - 97 days, 114 - 120 days, 115 - 120 days, 114 – 120 days, 90 – 95 days. All varieties belong to the Oryza sativa species. In the organic production model, farmers were trained in organic rice cultivation that followed the process of organic rice production issued by the Institute of Agricultural Sciences for Southern Vietnam (IAS, 2016). In Thoi Binh district, Ca Mau province, paddy was transplanted after 15 – 20 days after seedling. In other areas, paddy was sowed, and then paddy was constantly flooded with 5–7 cm depth from the soil surface at the four-leaf stage unstill one month before harvesting.

C. Sampling

Irrigation water, soil, and plants were sampled at the drought stage (around 20 days before harvest). Water samples were collected in 1 L plastic bottles, soil samples were collected at a depth of 0–10 cm, and plant samples were collected by chopping the above-ground portion of the plants. The collection of soil, water, and rice samples should accurately represent the entire production batch and target to detect residual chemicals thoroughly in high-risk areas (different from random sampling). The number of samples depends on the farming conditions of each household group and risk capacity. If farming conditions of the household is not good with high-risk possibility, samples need to be taken more. Each sample was collected from neighbouring households and then mixed as 1 sample for analysis. A total number of 40 samples was sampled and analyzed during five years (2017 - 2021).

D. Pesticide Analysis

The samples were sent for analysis at the labs designated by CU. Without washing, the samples were then extracted and analyzed using GC-MS/MS, as described by Braun *et al*. (2018), and LC-MS/MS described by Shah *et al*. (2015). The results from the analyses were reported in milligrams per kilogram (mg kg^{-1}).

E. Evaluating Criteria and Statistical Analysis

In this study, we use the criteria to evaluate the presence of pesticides as follows. Limit of detection (LOD) is the lowest quantity of an active ingredient that can be distinguished from the absence of active ingredients. Pesticide concentration (PC) (mg kg^{-1}) is the amount of pesticide's active ingredient per total weight of the sample. Progressive presence (PP) is the cumulative number of impressions from previous times. Frequency of occurrence (FOO) is the number of times that a pesticide is detected with a concentration higher than LOD, and contamination rate (CR) (%) is determined by the percentage of frequency of occurrence per the total number of samples. Microsoft Excel was used for data analysis and graphing.

III. Results and Discussion

A. Test Results of Prohibited Pesticides from Being Used in Organic Rice Production in Mekong Delta for Five Years (2017 – 2021)

In 2017, a total of 4 rice samples were collected in Cau Ngang district of Tra Vinh province in February. The test result showed that 3 samples were not contaminated with prohibited substances from being used in organic production. Only 1 sample was detected with difenoconazole (0.048 mg kg^{-1}) (Table III). Cau Ngang is a coastal district where paddy rice is cultivated intercropped with shrimp, just one crop for a year. That is a reason why fewer pesticide residues were detected in this area. This result agrees with Braun *et al.* (2019), who concluded that alternating rice-shrimp systems present a low-risk option in terms of food safety, which may bring additional benefits to this so far rather than a low-input system in the brackish water transition zone. However, some pesticides were applied in alternating rice-shrimp farming systems, either because they were still needed for maintaining the rice growth or as chemical pre-treatment of the rice seeds (pickling) (Braun *et al.*, 2019).

TABLE II: Pesticide Residues in Rice Fields of the Organic Rice Production Models in the Mekong Delta in 2017

Sample No.	Sampling sites	Sampling time	Active ingredients	LOD (mg kg^{-1})	RC (mg kg^{-1})	PP
1	CN-TV	14/02/2017	ND	-	-	-
2	CN-TV	14/02/2017	Difenoconazole	0.003	0.048	1
3	CN-TV	14/02/2017	ND	-	-	-
4	CN-TV	26/02/2017	ND	-	-	-

ND: not detected

In 2018, 1 rice sample was collected from Cau Ngang district of Tra Vinh province, and 3 samples were collected from Thoi Binh district of Ca Mau province from April to December (Table IV). Among 4 samples, only rice sample No. 6, collected in Thoi Binh district, was not contaminated with pesticides. The remaining 3 samples were contaminated with pesticides. Therein sample No. 5, collected in Cau Ngang district, was contaminated tricyclazole with a high concentration of 1.28 mg kg^{-1}. This result shows that tricyclazole is present even in the alternating rice-shrimp systems where farmers are aware of the negative effects of pesticide application on shrimp production in the subsequent season (Berg, 2001). However, the result agrees with Padovani *et al.* (2006) stated that tricyclazole (5-methyl-1,2,4-triazole(3,4-b) benzothiazole) is the most commonly used fungicide applied for controlling the rice blast disease in Asian countries. It also agrees with the study by Padovani *et al.* (2006) stated that tricyclazole provides rice with long-term protection during the whole growth process because of its stability and accumulation in soil. Because tricyclazole is relatively stable in water-soil systems, the potential environmental risk of tricyclazole is considered significant, so the WHO has classified tricyclazole as a moderately hazardous pesticide (WHO 2005). The other two samples (samples No. 7 - 8), collected in Thoi Binh district of Ca Mau province, were contaminated with 4 different active ingredients, including tricyclazole, propiconazole, tebuconazole, and difenoconazole. The residual concentration ranged from 0.085 mg kg^{-1} (difenoconazole and tricyclazole) to 0.309 mg kg^{-1} (difenoconazole).

TABLE III: Pesticide Residues in Rice Fields of the Organic Rice Production Models in the Mekong Delta in 2018

Sample No.	Sampling sites	Sampling time	Active ingredients	LOD (mg kg^{-1})	RC (mg kg^{-1})	PP
5	CN-TV	10/04/2018	Tricyclazole	0.005	1.28	1
6	TB-CM	19/12/2018	ND	-	-	-
7	TB-CM	19/12/2018	Propiconazole	0.007	0.188	1
			Tebuconazole	0.005	0.267	1
			Difenoconazole	0.003	0.309	2
			Tricyclazole	0.005	0.098	2
8	TB-CM	29/12/2018	Propiconazole	0.007	0.30	2
			Tebuconazole	0.005	0.097	2
			Difenoconazole	0.003	0.085	3
			Tricyclazole	0.005	0.085	3

ND: not detected

In 2019, A total of 14 samples (sample No. 9 - 22), including 1 water, 1 soil, and 12 rice samples, were tested in 6 districts of 4 provinces in the Mekong Delta from March to December (Table V). Therein, 4 samples collected in Tri Ton district of An Giang province, Vung Liem district of Vinh Long province, and Nga Nam district of Soc Trang province were not contaminated with prohibited substances in organic production. It is noted that these districts are located in the rice intensive-farming areas, where pest and disease pressure is very high, but no pesticide residue was detected. It proves that if farmers follow the organic production procedure, pesticide contamination risk in their farms would be low. Other 10 samples were contaminated with 7 active ingredients, including tricyclazole, chlorpyrifos-ethyl, isoprothiolane, propiconazole, hexaconazole, chlorfenapyr, and cypermethrin with the concentration ranging from 0.01 mg kg^{-1} (chlorpyrifos-ethyl and tricyclazole) to 0.71 mg kg^{-1} (tricyclazole).

TABLE IV: RESULTS OF ANALYSIS OF PESTICIDE RESIDUES IN RICE FIELDS OF THE ORGANIC RICE PRODUCTION MODELS IN THE MEKONG DELTA IN 2019

Sample No.	Sampling sites	Sampling time	Active ingredients	LOD (mg kg^{-1})	RC (mg kg^{-1})	PP
9	TT-AG	07/03/2019	Tricyclazole	0.005	0.71	4
			Propiconazole	0.007	0.60	3
10*	TT-AG	07/03/2019	ND	0.01	ND	0
11#	TT-AG	07/03/2019	Isoprothiolane	0.003	0.02	1
			Tricyclazole	0.005	0.07	5
12	VL-VL	22/05/2019	ND	-	-	-
13	NN-ST	22/05/2019	ND	-	-	-
14	NN-ST	27/05/2019	Tricyclazole	0.005	0.023	6
15	MX-ST	01/06/2019	Tricyclazole	0.005	0.022	7
			Cypermethrin	0.003	0.017	1
16	MX-ST	01/06/2019	ND	-	-	-
17	MX-ST	01/06/2019	Tricyclazole	0.005	0.024	8
18	TH-KG	01/08/2019	Tricyclazole	0.005	0.01	9
19	TT-AG	04/09/2019	Chlorpyrifos-ethyl	0.002	0.01	1
			Hexaconazole	0.007	0.015	1
			Tricyclazole	0.005	0.01	10
			Chlorfenapyr	0.008	0.063	1
20	UMT-KG	19/09/2019	Tricyclazole	0.005	0.015	11
21	TT-AG	23/12/2019	Chlorpyrifos-ethyl	0.002	0.022	2
			Isoprothiolane	0.003	0.021	2
			Tricyclazole	0.005	0.022	12
22	TT-AG	23/12/2019	Chlorpyrifos-ethyl	0.002	0.011	3
			Isoprothiolane	0.003	0.03	3
			Tricyclazole	0.005	0.031	13
			Hexaconazole	0.007	0.011	2

* Water sample; # Soil sample; ND: not detected

In 2020, a total of 6 rice samples (sample No. 23 - 28) were sampled from March to December. There were 4 samples collected in Tri Ton district of An Giang province, and 2 samples were collected from Thanh Tri district of Soc Trang province. All 6 samples were contaminated with the prohibited active ingredients (Table VI). The least contaminated sample was sample No. 25, collected in Tri Ton district of An Giang province, in which chlorpyrifos-ethyl was detected out of 854 substances, with a low concentration of 0.005 mg kg^{-1}. At the same time, sample No. 23, also collected from Tri Ton district of An Giang province, was most contaminated with 11 different active ingredients. This year, a total of 17 different active ingredients were detected, including tricyclazole, difenoconazole, chlorpyrifos-ethyl, isoprothiolane, propiconazole, azoxystrobin, hexaconazole, chlorfenapyr, cypermethrin, paclobutrazol, niclosamide, chlorfenson, fipronil, fipronil-desulfinyl, fenoxanil, metalaxyl & metalaxyl-M, and chlorfluazuron with concentration ranged from 0.005 mg kg^{-1} (chlorpyrifos-ethyl and fipronil-desulfinyl) to 2.1 mg kg^{-1} (metalaxyl & metalaxyl-M). Most of the contaminants were found in Tri Ton district of An Giang province.

TABLE V: RESULTS OF ANALYSIS OF PESTICIDE RESIDUES IN RICE FIELDS OF THE ORGANIC RICE PRODUCTION MODELS IN THE MEKONG DELTA IN 2020

Sample No.	Sampling sites	Sampling time	Active ingredients	LOD (mg kg^{-1})	RC (mg kg^{-1})	PP
23	TT-AG	20/3/2020	Azoxystrobin	0.005	0.22	1
			Chlorfenapyr	0.008	0.053	2
			Chlorpyrifos-ethyl	0.002	0.016	4
			Difenoconazole	0.003	0.56	4
			Fenoxanil	0.003	0.019	1
			Fipronil	0.002	0.0065	1
			Fipronil-desulfinyl	0.002	0.005	1
			Propiconazole	0.007	<0.02	4
			Isoprothiolane	0.003	0.014	4
			Hexaconazole	0.007	0.028	3
			Tricyclazole	0.005	0.16	14
24	TT-AG	31/03/2020	Chlorpyrifos-ethyl	0.002	0.005	5
			Difenoconazole	0.003	0.038	5
			Hexaconazole	0.007	0.02	4
			Isoprothiolane	0.003	0.01	5
			Metalaxyl & Metalaxyl-M	0.003	2.1	1
			Propiconazole	0.007	0.036	5
			Tricyclazole	0.005	0.021	15
25	TT-AG	27/11/2020	Chlorpyrifos-ethyl	0.002	0.01	6
26	TT-AG	27/11/2020	Azoxystrobin	0.005	0.036	2
			Chlorfenapyr	0.008	0.008	3
			Chlorfenson	0.003	0.11	1
			Cypermethrin	0.003	<0,01	2
			Difenoconazole	0.003	1.4	6
			Niclosamide	0.003	0.2	1
			Hexaconazole	0.007	1.2	5
			Tricyclazole	0.005	0.16	16
			Propiconazole	0.007	1.3	6
27	TT-ST	25/12/2020	Chlorfluazuron	0.008	0.2	1
			Chlorfenapyr	0.008	0.008	4
			Chlorpyrifos-ethyl	0.002	0.005	7
			Tricyclazole	0.005	0.01	17
28	TT-ST	25/12/2020	Azoxystrobin	0.005	0.015	3
			Chlorpyrifos-ethyl	0.002	0.017	8
			Difenoconazole	0.003	0.01	7
			Paclobutrazol	0.003	0.01	1
			Tricyclazole	0.005	0.01	18

The test result in 2021 is presented in Table VII. A total of 12 rice samples were tested. Therein, 2 samples, collected in U Minh Thuong district of Kien Giang province and Tri Ton district of An Giang province were not detected the prohibited substances (samples No. 34 and No. 40). Other samples were contaminated with active ingredients varied from 1 substance to 6 substances. Samples No. 37 and No. 39, both collected in Tri Ton district of An Giang province, were most contaminated with pesticides. It is clear to indicate that in the rice intensive-farming areas where the pressure of pests and diseases is high, farmers have broken the rules in organic production used pesticides on their farms when the organic methods (i.e. the methods using biological, mechanical, or chemical agents which are allowed to use in organic production according to organic standards) are failed or not effective. This induction could be cleared by a review of Teló *et al.* (2017), who reported that several studies found no residue of the fungicide difenoconazole in rice grains in China if that pesticide is applied 30 days before rice harvest, even with high rates of 90 g ha^{-1} and 135 g ha^{-1}. Thus, it is possible that farmers in the examined areas had applied fungicides too late, not more than 30 days until the sampling time. This reason is similar to the report by Nguyen & Van (2021) stated that the farmers did not follow organic production procedures to

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use organic-prohibited pesticides for their farms in recent years.

TABLE VI: RESULTS OF ANALYSIS OF PESTICIDE RESIDUES IN RICE FIELDS OF THE ORGANIC RICE PRODUCTION MODELS IN THE MEKONG DELTA IN 2021

Sample No.	Sampling sites	Sampling time	Active ingredients	LOD (mg kg-1)	RC (mg kg-1)	PP
29	UMT-KG	27/01/2021	Tricyclazole	0.005	0.01	19
			Isoprothiolane	0.003	0.022	6
30	UMT-KG	27/01/2021	Isoprothiolane	0.003	0.015	7
31	UMT-KG	27/01/2021	ND	-	-	-
32	UMT-KG	27/01/2021	Tricyclazole	0.005	<0.01	20
33	VL-VL	04/02/2021	Azoxystrobin	0.005	0.025	4
			Chlorpyrifos-ethyl	0.002	0.005	9
			Difenoconazole	0.003	0.027	8
34	TS-AG	14/04/2021	Hexaconazole	0.007	0.016	6
			Isoprothiolane	0.003	0.036	8
			Tricyclazole	0.005	0.01	21
35	VL-VL	27/05/2021	Azoxystrobin	0.005	0.25	5
			Chlorpyrifos-ethyl	0.002	0.005	10
			Difenoconazole	0.003	0.27	9
36	VL-VL	27/05/2021	Azoxystrobin	0.005	0.21	6
			Chlorpyrifos-ethyl	0.002	<0.005	11
			Difenoconazole	0.003	0.18	10
			Isoprothiolane	0.003	0.012	9
			Tricyclazole	0.005	<0.01	22
37	TT-AG	17/08/2021	Chlorfenapyr	0.008	0.015	5
			Cypermethrin	0.003	0.042	3
			Difenoconazole	0.003	0.033	11
			Paclobutrazol	0.003	0.054	3
			Propiconazole	0.007	0.036	7
			Tricyclazole	0.005	0.023	23
38	TT-AG	24/08/2021	Azoxystrobin	0.005	0.015	7
			Chlorpyrifos-ethyl	0.002	0.017	12
			Difenoconazole	0.003	0.01	12
			Isoprothiolane	0.003	0.01	10
			Tricyclazole	0.005	0.01	24
39	TT-AG	24/08/2021	Chlorfenapyr	0.008	0.01	6
			Cypermethrin	0.003	0.011	4
			Difenoconazole	0.003	<0.01	13
			Paclobutrazol	0.003	<0.01	2
			Propiconazole	0.007	<0.01	8
			Tricyclazole	0.005	0.01	25
40	TT-AG	14/09/2021	ND	-	-	-

ND: not detected

B. Summarizing the Presence of Tricyclazole and Other Prohibited Active Ingredients from Being Used in Organic Rice Production in the Mekong Delta Rice Fields during Five Years (2017 – 2021)

Through five years of research on the presence of tricyclazole and chemical active ingredients that are prohibited from being used in organic rice production, we found that the residual pesticides in rice crops came from two sources. The chemical pesticides used to prevent pests and diseases by farmers violated the organic production processes as discussed above. And another source was the pesticide residues that remained in the soil of the previous crops that could be realized based on the half-life (i.e. time for starting material to be reduced by 50%) of pesticides in the soil as discussed in the previous study (Duong *et al.*, 2022). There were 18 active ingredients present in 5 years (2017-2021), with the progressive presence and contamination rates are ordered as tricyclazole (25 times, 63%), difenoconazole (13 times, 33%), chlorpyrifos-ethyl (12 times, 30%), isoprothiolane (10 times, 25%), propiconazole (8 times, 20%), azoxystrobin (7 times, 18%), hexaconazole (6 times, 15%), chlorfenapyr (6 times, 15%), cypermethrin (4 times, 10%), paclobutrazol (2 times, 5%), tebuconazole (2 times, 5%) and seven remaining ingredients including niclosamide, chlorfenson, fipronil, fipronil-desulfinyl, fenoxanil, metalaxyl & metalaxyl-M, and chlorfluazuron with having the same progressive presence and contamination rate of 1 time and 2.5%, respectively (Fig. 2). Therein, 8 insecticides, 7 fungicides, 1 snail killer, 1 seed treatment, and 1 growth regulator (Table VII). This result is similar to what was reported by IRRI (2018), stating that fungicides and insecticides are frequently used to control brown planthopper, brown spot or rice blast, which are the most common pests and diseases for paddy rice crops.

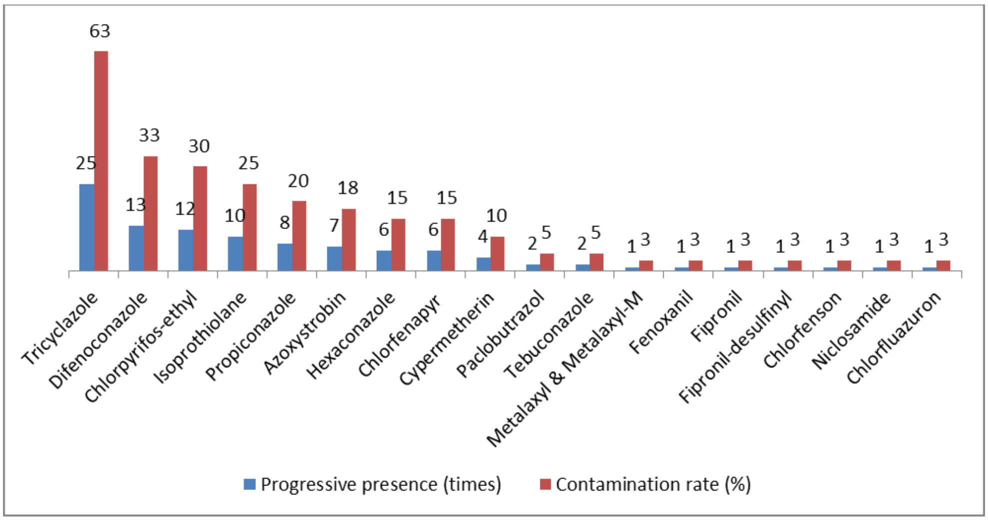

Fig. 2. The progressive presence and contamination rate of residual pesticides in the Mekong Delta rice fields were tested during five years (2017 – 2021).

From this study, tricyclazole was identified in the Mekong Delta with the highest prevalence with the highest rate of 63%. At the same time, a report on tricyclazole residue study in rice in India stated that residues of tricyclazole were detected in 45% of the samples, of which 31.37% contained it above the maximum residue level (MRL, 0.01 mg kg^{-1}) fixed by Food Safety and Standards Authority of India (Naik *et al.*, 2020). Tricyclazole is an active ingredient that farmers believe is effective against fungal diseases, especially rice blasts. It is similar to the report of Peterson (1990), who stated that tricyclazole is a unique fungicide to control *Pyricularia oryzae* disease in rice. Compared the result of this study to previous studies on pesticides assessment of paddy rice in Mekong Delta, there were five pesticides, including propiconazole, hexaconazole, isoprothiolane, cypermethrin, and fipronil, are matched with the study by Berg (2001), seven pesticides, including propiconazole, hexaconazole, isoprothiolane, tricyclazole, cypermethrin, fipronil, and chlorpyrifos are matched with the report by Berg & Tam (2012), and seven pesticides, including chlorpyrifos, fipronil, difenoconazole, propiconazole, hexaconazole, isoprothiolane, and azoxystrobin, identified in this study are also reported by Braun *et al.* (2019).

TABLE VII: SUMMARIZING THE PRESENCE OF CONTAMINATED ACTIVE INGEDIENTS IN THE MEKONG DELTA RICE FIELDS ACCORDING TO TIME AND LOCATIONS

No.	Active ingredients	Usages	Present years	Present sites
1	Tricyclazole	Fungicide	2018, 2019, 2020, 2021	CN-TV, TB-CM, TT-AG, TS-AG, TH-KG, UMT-KG, NN-ST, TT-ST, MX-ST VL-VL
2	Difenoconazole	Seed treatment	2017, 2018, 2020, 2021	CN-TV, TB-CM, TT-AG, TT-ST, VL-VL
3	Propiconazole	Insecticide	2018, 2019, 2020, 2021	TB-CM, TT-AG
4	Isoprothiolane	Fungicide	2019, 2020, 2021	TT-AG, TS-AG, UMT-KG, VL-VL
5	Chlorpyrifos-ethyl	Fungicide	2019, 2020, 2021	TT-AG, TT-ST, VL-VL
6	Hexaconazole	Fungicide	2019, 2020, 2021	TT-AG, TS-AG
7	Chlorfenapyr	Fungicide	2019, 2020, 2021	TT-AG, TT-ST
8	Cypermethrin	Insecticide	2019, 2020, 2021	TT-AG, MX-ST
9	Azoxystrobin	Insecticide	2020, 2021	TT-AG, TT-ST, VL-VL
10	Paclobutrazol	Growth regulator	2020, 2021	TT-AG, TT-ST
11	Tebuconazole	Fungicide	2018	TB-CM
12	Niclosamide	Snails killer	2020	TT-AG
13	Chlorfenson	Insecticide	2020	TT-AG
14	Fipronil	Insecticide	2020	TT-AG
15	Fipronil-desulfinyl	Insecticide	2020	TT-AG
16	Fenoxanil	Insecticide	2020	TT-AG
17	Metalaxyl & metalaxyl-M	Fungicide	2020	TT-AG
18	Chlorfluazuron	Insecticide	2020	TT-AG

Classifying according to the present time, they can be divided into 4 groups. The 4-year presence group includes tricyclazole, difenoconazole, and propiconazole. Therein, tricyclazole was most prominent, which was present in 10 districts of 6 provinces, while difenoconazole was present in only 5 districts of 5 provinces, and propiconazole was present in 2 districts of 2 different provinces. The 3-year presence group includes isoprothiolane, chlorpyrifos-ethyl, hexaconazole, chlorfenapyr, and cypermethrin. Therein, isoprothiolane was present in 4 districts of 3 provinces, chlorpyrifos-ethyl was present in 3 districts of 3 provinces, and others were just present in 2 districts. The 2-year presence group includes azoxystrobin and paclobutrazol, in which azoxystrobin was present in 3 districts of 3 provinces and paclobutrazol was present in 2 districts of 2 provinces. The 1-year presence group includes 8 active ingredients which were present only district, including tebuconazole, niclosamide, chlorfenson, fipronil, fipronil-desulfinyl, fenoxanil, metalaxyl & metalaxyl-M, and chlorfluazuron (Table VII).

IV. CONCLUSION

To evaluate the presence of pesticide residues that are prohibited used in organic rice production, the collected rice samples in six provinces in Mekong Delta were analyzed by LC-MS/MS and GC-MS/MS methods. We found 18 residual active ingredients in the soil and rice samples. Based on the contamination rate, we divide them into three groups. The high presence group has a contamination rate greater than 50%, including only tricyclazole (63%). The medium presence group is those with contamination rate greater from 10% to 50% including difenoconazole (33%), Chlorpyrifos-ethyl (30%), isoprothiolane (25%), propiconazole (20%), azoxystrobin (18%), hexaconazole (15%), chlorfenapyr (15%), cypermethrin (10%). The low presence group is those with a contamination rate of less than 10%, including paclobutrazol (5%), tebuconazole (5%) and the seven remaining ingredients niclosamide, chlorfenson, fipronil, fipronil-desulfinyl, fenoxanil, metalaxyl & metalaxyl-M, and chlorfluazuron with the same contamination rate (2.5%). Classifying according to the number of years of presence, they can be divided into 4 groups. The 4-year presence group includes tricyclazole, difenoconazole, and propiconazole. The 3-year presence group includes Chlorpyrifos-ethyl, isoprothiolane, hexaconazole, chlorfenapyr, and cypermethrin. The 2-year presence group includes azoxystrobin and paclobutrazol. The 1-year presence group includes 8 active ingredients, including tebuconazole, niclosamide, chlorfenson, fipronil, fipronil-desulfinyl, fenoxanil, metalaxyl & metalaxyl-M, and chlorfluazuron. Among 18 detected active ingredients, there were 8 substances used for disease control, 8 substances used for insecticides, 1 substance used for killing snails, and 1 substance used as growth regulation. The research result is valuable for building organic rice material zones because it helps to reduce the time, risks and analysis costs. Hereby, we suggest the governmental and non-governmental organizations increase financial support for AOI to organize and build the linkage model of the rice value chain according to organic standards to minimize the use of pesticides which is harmful to the environment and consumers' health.

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CONFLICT OF INTEREST

The authors declare that they do not have any conflict of interest.

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