

Pollution by wastewater from aquaculture in some coastal zone in the Mekong Delta, Vietnam

¹Luom T. Thanh, ²Huy B. Le, ³Quang M. Dinh, ²Hoan X. Nguyen, ²Hiep T. Dang, ²Thanh M. Le, ¹Ngoc T. H. Dang

¹ Faculty of Natural Resource and Environment, University of Kien Giang, Kien Giang, Viet Nam; ² Ho Chi Minh City University of Food Industry, Ho Chi Minh City, Viet Nam;

³ Department of Biology, School of Education, Can Tho University, Can Tho, Vietnam.
Corresponding author: Q. M. Dinh, dmquang@ctu.edu.vn

Abstract. This study assessed the current aquaculture, especially industrial shrimp farming, causing soil and water pollution. Also, the mass death of shrimp and fish on a large area in the Mekong Delta, caused by wastewater and sludge with chemical residues was examined in the present study. All water samples analyzed above-contained cypermethrin (pesticide) at concentrations greater than 0.01 ppb, which could cause mortality of 76.2% or higher, after 35 days (e.g. caused by liver and kidney necrosis), in fish and shrimp populations. Tra Vinh was used as a case study for this assessment. Accordingly, farming shrimp activities affected soil degradation, such as acidity, salinization or spread of toxic elements (Al^{3+} , Fe^{2+} , H^+) into the water. Water in most canals was contaminated. NH_3 , chemical oxygen demand, chromaticity and suspended solids were higher than the allowed standard. Thus, aquaculture wastewater could be hazardous.

Key Words: aquaculture, cypermethrin, liver and kidney necrosis, water pollution, Mekong Delta.

Introduction. The Mekong Delta (VMD) is one of the largest fishing grounds in Vietnam. With the advantage of a coastline of over 780 km and 297,000 km², the marine exclusive economic zone has brought abundant fishery resources to the region (Le 2006). The estimated exploitation capacity at VMD is 830,000 tons year⁻¹, accounting for 40.6% of total output (General Statistics Office 2017). Besides, the number of aquatic species, especially fish species is very rich and diverse. According to research conducted by Tran et al (2013), in VMD there are about 322 species belonging to 77 families, distributed from freshwater to saltwater. There are many species with high nutritional and economic value (Nguyen 2000). Therefore, fishing in VMD is very developed with many types of fishing gear and boats (Nguyen & Nguyen 2010). Aquaculture has been the most important economic sector in the Mekong Delta since 2000. The recent rapid increase of the combined and specialized aquaculture in fresh and saltwater areas has improved farmers' livelihoods, used rural labour and increased exports, whose value reached 1 billion USD in 2002, 1.5 billion in 2005 and about 6 billion, currently. According to the General Statistics Office (2017), the GDP of fisheries reached more than 190,000 billion VND, accounting for 3.43% of the entire economy and more than 23% of the whole agricultural sector. The Mekong Delta contributes more than 74% of aquaculture production nationwide, where shrimp farming is dominant. Simultaneously, environmental pollution in river basins, canals and other surface waters pollution is increasing (Huy et al 2012). This phenomenon causes the mass death of shrimp and fish in the entire farming area of the Vam Co Dong river basin, Vi Thanh River, Ong Doc river, Bay Hap river, downstream of Tien river, Hau river, in the recent years.

According to FAO (2003), uneaten food is the main cause of water pollution in these areas. Water from ponds that are directly discharged into the environment is also a cause of water pollution (Tran 2016). In addition, the improper use of pesticides, improper storage, and improper packaging destruction are the causes of pesticide residues in soil and water (Pham et al 2011). A study by Bui & Nguyen (2014) showed

that in Ca Mau, about 83.4% of shrimp farming households pump sludge into storage and about 16.6% of shrimp farming households discharge sludge directly into rivers and canals. In Can Tho, on average, each farmer sprays pesticides five times in a rice crop and the concentrations are 2-3 times higher than recommended (Nguyen et al 2020). The amount of wastewater and waste from intensive farming facilities needs to be controlled (Ministry of Agriculture and Rural Development 2013; Le et al 2018). Some of the shrimp farming pollutants are BOD5 (biological oxygen demand), chemical oxygen demand (COD), N, P, and suspended solids (SS) (Le et al 2012), contaminating sludge in ponds of the Mekong Delta (Chau et al 2012), essentially with high P and N residues (Yang et al 2004). Not only harmful to aquatic species, but pesticide residues also adversely affect human health (Bui & Lam 2010). This research aimed to provide an objective and thorough insight into the application of aquaculture in the Mekong Delta, as the basis for the management planning.

Material and Method

Sample collection. According to Huy (2000), 600 Remote Sensing & Geographic Information Systems (RS & GIS) image samples were collected from a geospatial area at the whole province level, with a map scale of 1:100,000, at a critical site level using a map scale of 1:25,000 and at the lower section of the Mekong River, using a map scale of 1:500,000. A JPS locator was included. Images were stored in the "Cloud Electronics".

Sample analysis. According to the sampling scheme, sample specimens were preserved and analyzed according to TCVN 2005-2013 Vietnamese National Standard Method. The number of samples (between 30 and 600) depended on the geospatial regions and on the environmental components and ecosystems used as input into EXCEL or SPSS. In the experiment design, 4 repetitions were considered. A control sample was always used and the coefficient of variance (CV%) was calculated, to have a basis for a correct evaluation. Nine water and 13 sediment samples were collected in Soc Trang and Bac Lieu provinces, then used to analyze the pesticide indicators and analytical methods. Seven pesticides were selected for residue analysis in water and sludge samples (Table 1). These drugs are commonly used in the Mekong Delta (except endosulfan) and toxic to aquatic animals. Pesticide residues in water samples were extracted from samples with dichloromethane and determined by gas chromatography with an ECD probe. Pesticide residue in the sludge sample was removed from the sample with a mixture of acetone: dichloromethane: petroleum ether in the ratio of 4:3:3, then cleaned through a silica gel column and determined by gas chromatography of the ECD probe.

Results

The pesticide residues in the water samples. The survey results of pesticide residues in water and sediment samples are shown in Table 1.

Table 1
The LOD (detection limit) and LOQ (determination limit)

Pesticide chemical	Chemical pesticide group	Water		Sediment sample	
		LOD ($\mu\text{g L}^{-1}$)	LOQ ($\mu\text{g L}^{-1}$)	LOD (mg kg^{-1})	LOQ (mg kg^{-1})
Cypermethrin	Cucumber	0.001	0.005	0.001	0.005
Permethrin	Cucumber	0.001	0.005	0.001	0.005
Deltamethrin	Cucumber	0.001	0.005	0.001	0.005
Endosulfan	Organic chloride	0.001	0.005	0.001	0.005
Chlorpyrifos - Ethyl	Organic phosphor	0.001	0.005	0.001	0.005
Profenofos	Organic phosphor	0.010	0.050	0.010	0.050
Fipronil	Other group	0.001	0.005	0.001	0.005

LOD ($\mu\text{g L}^{-1}$); LOQ ($\mu\text{g L}$); LOD (mg kg^{-1}); LOQ (mg kg^{-1}).

Pesticide residues in water, Soc Trang province, Mekong Delta were presented in Table 2.

Table 2

Analysis results of pesticide residues in water samples ($\mu\text{g L}^{-1}$)

Sample	Pesticide residues ($\mu\text{g L}^{-1}$)						
	CP	PM	DLM	ES	CPS	PRF	FP
ST01	0.032	0.113	<LOD	<LOD	0.018	<LOD	<LOD
ST02	0.025	0.098	<LOD	<LOD	<LOD	<LOD	<LOD
ST03	0.025	<LOD	<LOD	<LOD	<LOD	<LOD	0.048
ST05	0.026	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ST06	0.025	<LOD	<LOD	<LOD	0.017	<LOD	0.007
ST07	0.016	<LOD	<LOD	<LOD	0.014	<LOD	<LOD
ST08	0.026	<LOD	<LOD	<LOD	<LOD	<LOD	0.020
ST09	0.030	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

CP-Cypermethrin; PM- Permethrin; DLM-Deltamethrin; ES-Endosulfan; CPS-Chlorpyrifos; PRF-Profenofos; FP-Fipronil; LOD-limit of detection.

The cypermethrin. The cypermethrin residues in water samples was 0.016-0.032 $\mu\text{g L}^{-1}$. The highest and lowest value of cypermethrin residues was found at at ST01 (0.032 $\mu\text{g L}^{-1}$) and ST07 (0.016 $\mu\text{g L}^{-1}$). The average value of cypermethrin residue was 0.026 $\mu\text{g L}^{-1}$ (Figure 1).

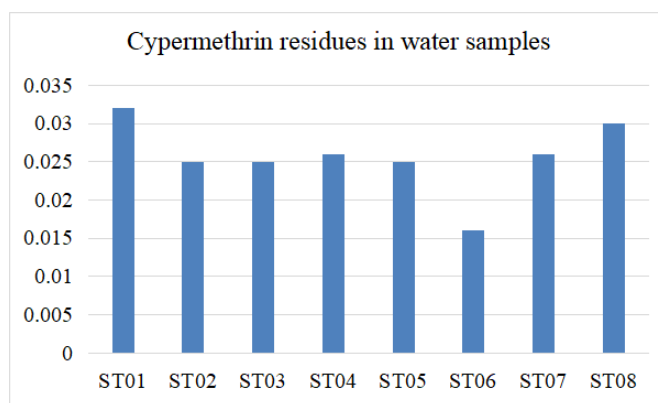


Figure 1. Cypermethrin residues in water samples.

The permethrin. The permethrin residues of waster samples was 0.098-0.113 $\mu\text{g L}^{-1}$. Permethrin residues was 0.113 at ST01 and 0.098 $\mu\text{g L}^{-1}$ at ST02 but not found from other sites. The average value of cypermethrin residues was 0.106 $\mu\text{g L}^{-1}$ (Figure 2).

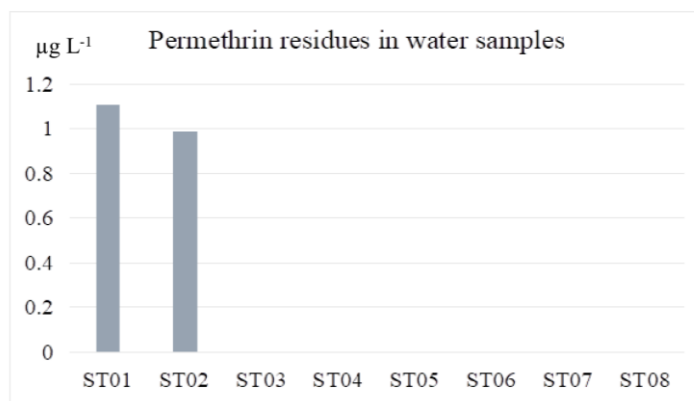


Figure 2. Permethrin residues in water samples.

The chlorpyrifos. The chlorpyrifos residues was 0.014-0.018 $\mu\text{g L}^{-1}$, reaching the

highest value at ST01 ($0.018 \mu\text{g L}^{-1}$), followed closely by ST06, and lowest at ST07 ($0.014 \mu\text{g L}^{-1}$). The chlorpyrifos residue was not found at other sites. The average value of chlorpyrifos residues was $0.016 \mu\text{g L}^{-1}$ (Figure 3).

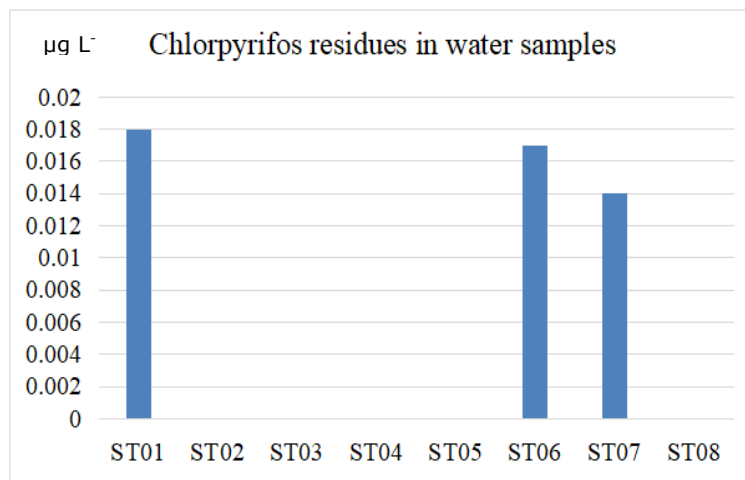


Figure 3. Chlorpyrifos residues in water samples.

The fipronil. The fipronil residue was $0.007\text{-}0.048 \mu\text{g L}^{-1}$. The highest and lowest value of this residue was $0.048 \mu\text{g L}^{-1}$ at ST03 and $0.007 \mu\text{g L}^{-1}$ at ST06, respectively. The average value of fipronil residues was $0.025 \mu\text{g L}^{-1}$ (Figure 4).

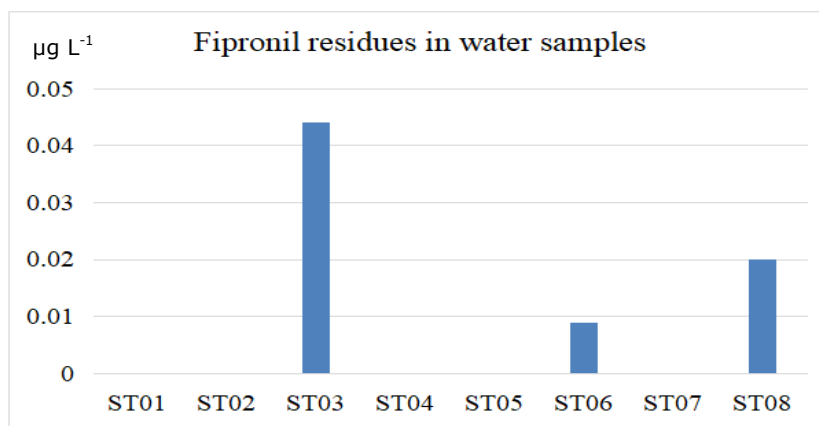


Figure 4. Fipronil residues in water samples.

Deltamethrin, endosulfan, and profenofos were not detected in all water samples.

Pesticide residues in the sludge in Bac Lieu and Soc Trang provinces. Among 13 sludge samples, only two residues of plant protection chemicals were detected, namely cypermethrin (23.08%) and permethrin (15.38%) (Table 3).

Table 3

Analysis results of pesticide residues in sludge samples

Name, code	Pesticide residues (mg kg^{-1})						
	CP	PM	DLM	ES	CPS	PRF	FP
BL1 (Soc Trang sludge 04)	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
BL (Soc Trang sludge 09)	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
BL (Soc Trang sludge 05)	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ST 2B	<LOD	0.009	<LOD	<LOD	<LOD	<LOD	<LOD
ST 3B	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ST 4B	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

Name, code	Pesticide residues ($mg\ kg^{-1}$)						
	CP	PM	DLM	ES	CPS	PRF	FP
ST 5B	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ST 6B	<LOQ	0.018	<LOD	<LOD	<LOD	<LOD	<LOD
ST 8B	0.005	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD
ST 9B (Dried sludge)	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
ST 10B (Sludge rice-shrimp)	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Bac Lieu sludge 01	0.108	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Bac Lieu sludge 02	0.007	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

CP-Cypermethrin; PM- Permethrin; DLM-Deltamethrin; ES-Endosulfan; CPS-Chlorpyrifos; PRF-Profenofos; FP-Fipronil; LOD: limit of detection.

The cypermethrin. The approximate value of cypermethrin in sludge samples was 0.005-0.108 $\mu g\ L^{-1}$. Cypermethrin residues showed the highest value of 0.108 $\mu g\ L^{-1}$ at ST10B and the lowest avalue of 0.005 $\mu g\ L^{-1}$ at ST8B. The average value of cypermethrin residues was 0.04 $\mu g\ L^{-1}$ (Figure 5).

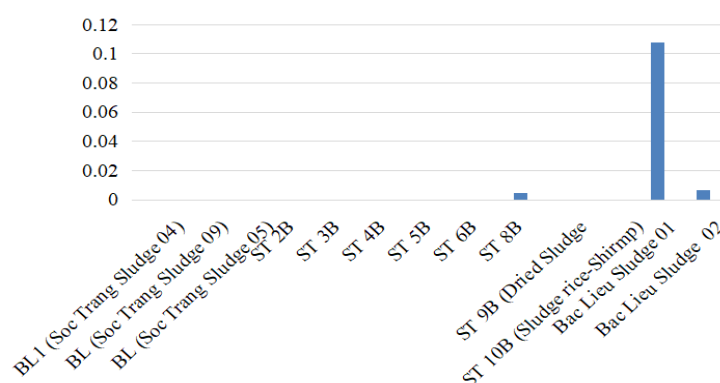


Figure 5. Cypermethrin residues in sludge samples.

The permethrin. The permethrin in sludge samples was 0.009-0.018 $\mu g\ L^{-1}$. Permethrin was found in the highest value of 0.018 $\mu g\ L^{-1}$ at ST6B and the lowest of 0.009 $\mu g\ L^{-1}$ at ST2B. The average value of permethrin residues was 0.135 $\mu g\ L^{-1}$ (Figure 6).

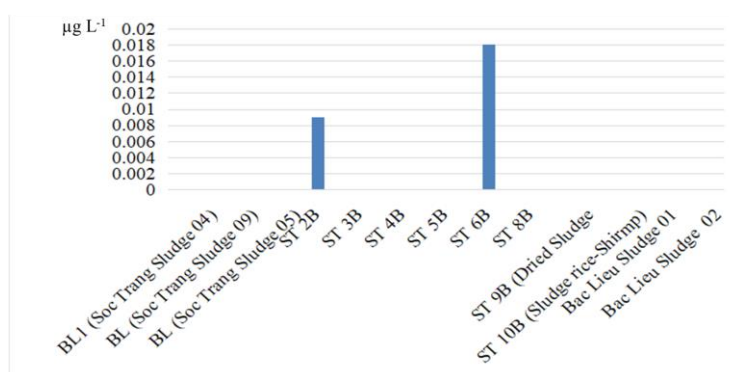


Figure 6. Permethrin residues in sludge samples.

Pollution caused by industrial shrimp farming activities in Tra Vinh province. The concentration of iron was 0.08-19.18%. The concentration of iron displayed the lowest value of 0.08% in Long Huu – Duyen Hai, and highest value of 19.18% in Long Toan – Duyen Hai district (Figure 7).

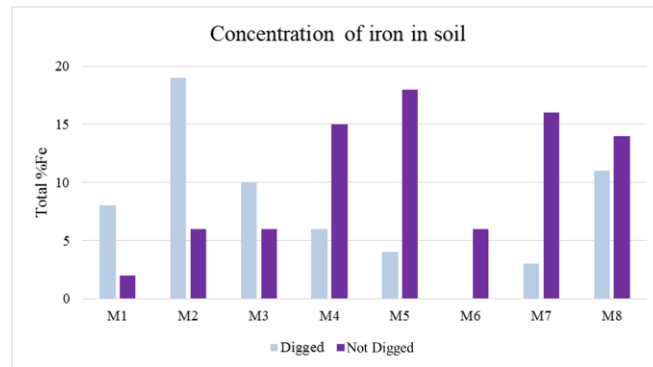


Figure 7. Concentration of iron in soil Tra Vinh province (M1: Hiep My; M2: Long Toan-Giong Trom 1; M3: Long Toan-Giong Trom 2; M4: Long Huu-Phuoc Tri 1; M5: Long Huu-11 Hamlet; M6: Long Huu-12 Hamlet; M7; Long Khanh-Cai Doi 1; M8: Long Khanh-Cai Doi 2).

The concentration of aluminum. Ion Al^{3+} in land ranged from 0.83 to 144.77 mg 100 g^{-1} . The value highest of Al^{3+} was 144.77 mg in Long Khanh – Duyen Hai district, and the lowest was 0.83 mg in Long Toan - Giong Trom (Figure 8).

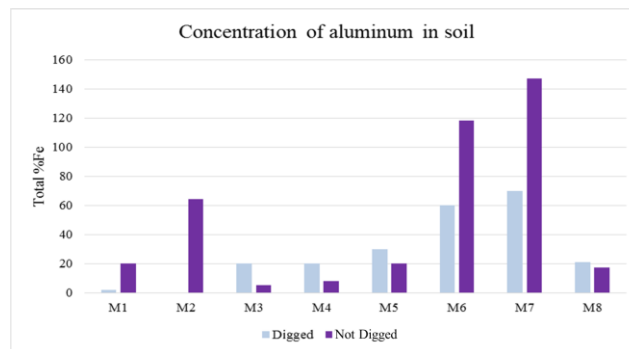


Figure 8. Concentration of aluminum in soil Tra Vinh province (M1: Hiep My; M2: Long Toan-Giong Trom 1; M3: Long Toan-Giong Trom 2; M4: Long Huu-Phuoc Tri 1; M5: Long Huu-11 Hamlet; M6: Long Huu-12 Hamlet; M7; Long Khanh-Cai Doi 1; M8: Long Khanh-Cai Doi 2).

Humus acid concentration and organic materials. The result showed that OM displayed a value of 1.48–3.61%. At positions 3 and 4 (Long Thanh, Long Toan), and 14 (Dinh Cuu, Long Khanh) chemical content was very high, ranging from 3.14, 3.13, 3.13, and 3.61%, respectively (Figure 9).

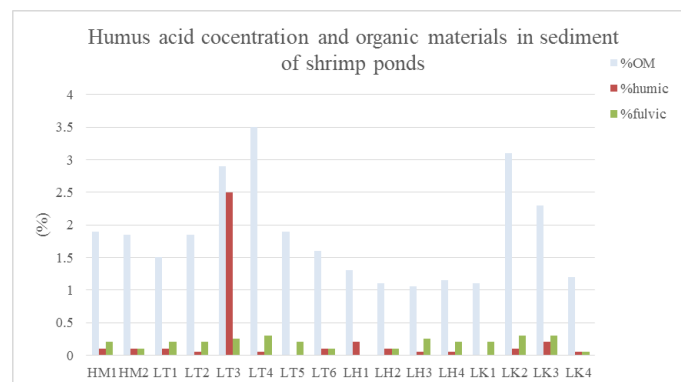


Figure 9. Humus acid concentration and organic materials in the sediment of shrimp ponds Tra Vinh province (Two symptoms are HM1 and HM2 in My Hiep commune; in Long Toan commune: LT1, LT2, LT3, LT4, LT5, LT6; in Long Huu commune: LH1, LH2, LH3, LH4 and Long Khanh commune: LK1, LK2, LK3, LK4).

Water environment pollution

The concentration of suspended solids. SS in two seasons altered below the permitted standard (80 mg L^{-1}). The SS in the dry season alters more than in the rainy season (Figure 10). In the dry season, SS was more than the standard limits (94 mg L^{-1} in Cau Ngang to 377 mg L^{-1} in Cai Doi – Duyen Hai), while in the rain season, it was just only from 149 mg L^{-1} in 11 hamlets, Duyen Hai.

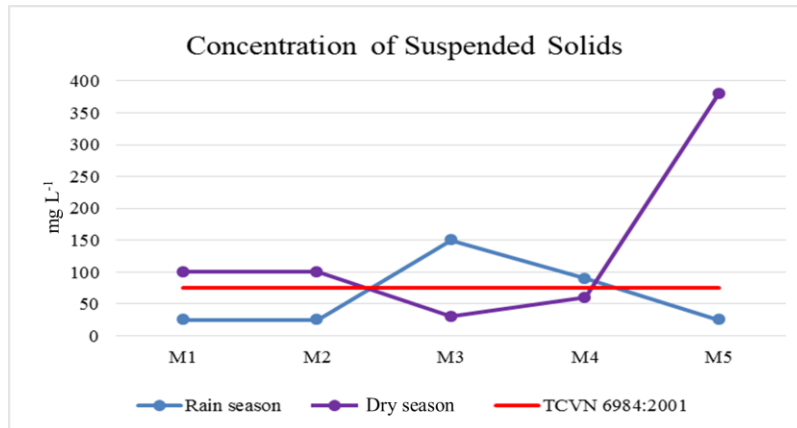


Figure 10. Concentration of suspended solids in water environment Tra Vinh province (M1: My Quy-Cau Ngang; M2: Gong Trom-Duyen Hai; M3: 11 Hamlet-Duyen Hai; M4: 12 Hamlet-Duyen Hai; M5: Cai Doi-Duyen Hai).

Chromaticity. In two seasons, chromaticity in canals consistently exceeded the allowed standard (50 Pt-Co). In the rainy season, the highest chromaticity was 475 Pt-Co at 11 hamlets, Duyen Hai, and in the dry season, the highest chromaticity was 1690 Pt-Co in Cai Doi, Duyen Hai (Figure 11).

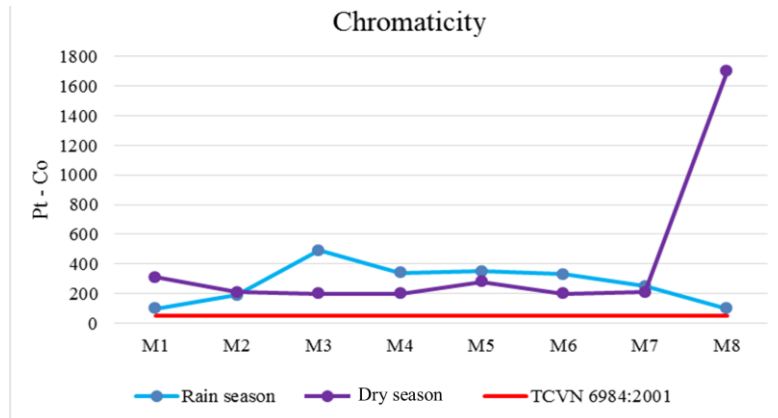


Figure 11. Chromaticity in water environment Tra Vinh province (M1: My Quy-Cau Ngang; M2: Gong Trom-Duyen Hai; M3: 11 Hamlet-Duyen Hai; M4: 12 Hamlet-Duyen Hai; M5: KX1 Dinh Cu-Duyen Hai; M5: KX2 Dinh Cu-Duyen Hai; M7: KX1 Cai Doi-Duyen Hai; M8: KX2 Cai Doi-Duyen Hai).

The concentration of chemical oxygen demand. COD in the rainy season ranged from 160 mg L^{-1} in My Quy – Cau Ngang) to 465 mg L^{-1} in Cai Doi, Duyen Hai, displaying over 3–8 times than the standard threshold ($<60 \text{ mg L}^{-1}$). In the dry season, COD ranged from 68.6 mg L^{-1} in My Quy – Cau Ngang to 98.6 mg L^{-1} in KX2 12 Hamlet - Duyen Hai) (Figure 12).

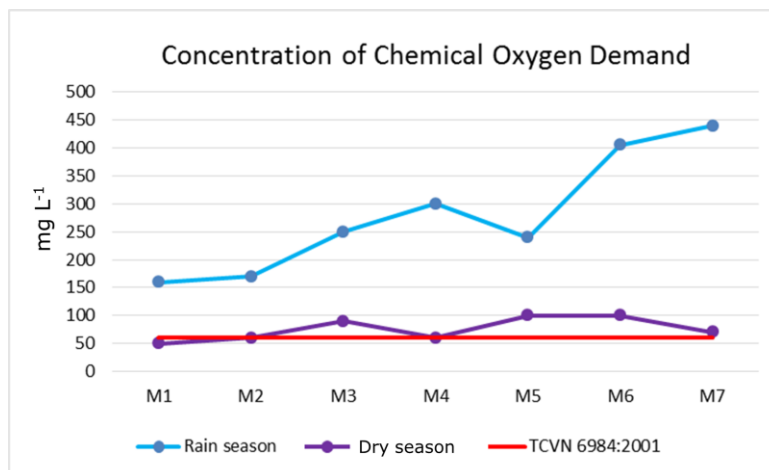


Figure 12. Concentration of chemical oxygen demand in water environment Tra Vinh province (M1: KX1 My Quy-Cau Ngang; M2: KX2 My Quy-Cau Ngang; M3: KX2 Gong Trom-Duyen Hai; M4: KX2 Phuoc Tri-Duyen Hai; M5: KX2 11 Hamlet-Duyen Hai; M6: KX2 12 Hamlet-Duyen Hai; M7: KX2 Cai Doi-Duyen Hai).

Concentration of N-NH₃. In the rainy season, ammonia content in most canals was over the permitted standard and ranged from 1.8 mg L⁻¹ in Giong Trom, Duyen Hai to 4.7 mg L⁻¹ in Cai Doi, Duyen Hai. This concentration ranged 1.11 mg L⁻¹ in My Quy – Cau Ngang to 3.19 mg L⁻¹ in KX2 Dinh Cu - Duyen Hai in the dry season (Figure 13).

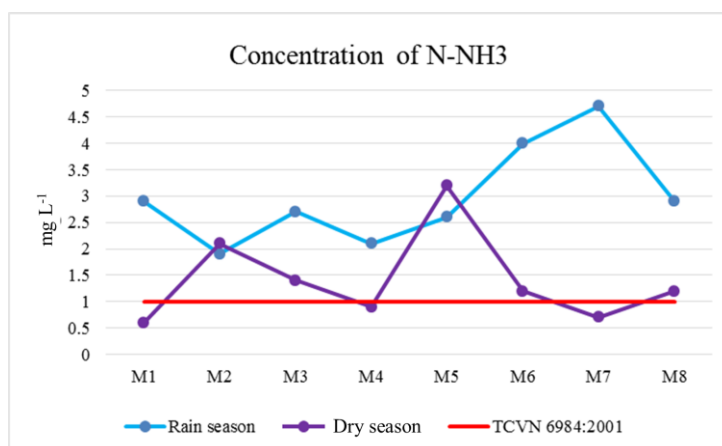


Figure 13. Concentration of N-NH₃ in water environment Tra Vinh province (M1: KX2 My Quy-Cau Ngang; M2: KX1 Gong Trom-Duyen Hai; M3: KX2 Phuoc Tri-Duyen Hai; M4: KX1 11 Hamlet-Duyen Hai; M5: KX1 Dinh Cu-Duyen Hai; M5: KX2 Dinh Cu-Duyen Hai; M7: KX 1 Cai Doi-Duyen Hai; M8: KX2 Cai Doi-Duyen Hai).

Discussion. It can be seen that two types of plant protection chemicals, Cypermethrin and Permethrin, were detected mainly in both water and mud samples. Cypermethrin was a synthetic pyrethroid insecticide (Huy 2017) with a chemical structure similar to pyrethrins, a natural insecticide derived from plants in the chrysanthemum family. If pyrethrins were unstable compounds and did not last long enough to kill insects, synthetic pyrethroid compounds, on the other hand, were durable and persisted for days or weeks sufficient to kill insects. This was why cypermethrin was widely used and found in the environment; cypermethrin kills insects by disrupting the normal function of the nervous system, leaving the nerves in a state of constant stimulation, which leads to convulsions and ultimately death. In addition, cypermethrin also affects other functions such as cell energy production, transport of metal atoms, and muscle contraction. In addition, cypermethrin also reduces the body's resistance, reproductive effects, mutations, and cancer (Brown 2006; Cox 1996). Effects of cypermethrin, at different concentrations, on survival and necrosis of the liver organs (Thi 2002), as well as on the

pancreas, in tiger shrimp cultured in experimental conditions in a closed house, were evaluated. Results showed that cypermethrin at 0.05 ppb caused 100% mortality after ten days. The other tested concentrations of cypermethrin were 0.01, 0.001 and 0.0001 ppb, causing mortality after 35 days of 76.2, 45.2 and 30.6%, respectively.

Particularly for shrimp farming in Tra Vinh province, after two years of research in two districts, Duyen Hai and Cau Ngang, the Sub-Institute of Fisheries Economics and Management in HCMC (2002) found certain amounts of toxins in shrimp ponds: 1-The content of Al^{3+} exchanged in the soil in the dry season was significantly greater compared to the rainy season, ranging from 12.43 to 234.62 mg g^{-1} of soil. The difference in Al^{3+} content exchanged between rainy and dry seasons in most study sites was very large. The process of leaching a large amount of Al^{3+} toxins into the pond occurs at the beginning of the rainy season. Therefore, at the start of the rainy season, shrimp farming areas need to be tightly controlled and appropriate bunker management measures are required to avoid shock and mortality, especially in the Duyen Hai district because there was a low shape, lying on salty alum soil close to the sea. Therefore, during the rainy season, the influence of alum ions (Al^{3+} , H^+ , Fe^{2+}) in the Duyen Hai district has a solid and profound impact on aquaculture (especially Long Toan commune). For this reason, it was necessary to have appropriate management measures for aquaculture in the late dry season and early rainy season when leaching occurs. The pH value ranged from 6.4 to 7.0; Particularly in the Duyen Hai district, pH fluctuates in a quite wide range, from 3.2 to 8.3, and soils were from very acidic to slightly alkaline. The EC (mS/cm) conductivity of the soil at the dry wet season entry locations was also very high compared to the rainy season due to the phenomenon of capillary water evaporation that makes the topsoil more heavily saline. EC in the dry season ranges from 182.8 to 420 mS/cm, 6-50 times higher than in the rainy season. Thus, rain plays a significant role in the process of salting and washing the soil alum. Therefore, it was necessary to have appropriate measures to take advantage of rainwater in washing alum and saline soils and avoid toxins, caused by alum and salinity once washed away, toxic to affecting the regional ecosystem surroundings and the area receiving this wash water source. The concentration of As in the soil in the two surveyed districts ranged from 7.74 to 38.72 ppm (average 20 ppm), still in the natural fluctuation range of the soil (no signs of arsenic contamination). Hg ranged from 0.08 to 0.14 ppm (average 0.11 ppm), in the normal range of the soil and is still very far behind the permitted standards. Cu ranged between 23 and 42 ppm (average 32.4 ppm) was close to the allowable threshold (36 ppm, according to Dutch standards).

Na^+ ranged between 5,041 and 14,620 ppm (average 9,525 ppm). The high Na values were mainly in soil samples collected from the edges of shrimp ponds. This is an important ion that plays a role in causing soil salinity, when it accumulates in the soil, it pushes the acidic ions such as Al^{3+} and H^+ out of the soil glue, making the soil no longer acidic but neutral or alkaline. The total content of iron in the dry season fluctuated in the range of 6.60-22.77% Fe_2O_3 , and in most places it was 1.1 to 138.6 times higher than in the rainy season. Total iron content increased because the mud bottom was transported to the shore and was also due to the alum effervescence into the dry season. Dry salt occurs with water evaporation, causing alum in the lower layer along the capillary tubes to be transferred to the upper layer. In addition, in dry season also occurs the intense oxidation of iron compounds in the mineral form.

The process of changing the strong iron content from Fe^{2+} to Fe^{3+} between the dry and rainy seasons takes place in the Duyen Hai district (coastal). In the dry season, alum effervescence occurs (turning from potential alum to active alum), and in the rainy season, alum toxicity will be washed out from the pond banks to the pond. Although the iron content in the soil is lower than in some other locations, the exploitation of the soil was not good, leading to solid humification.

Conclusions. The potential for aquaculture in the Mekong Delta was huge, diverse, and with high economic efficiency, but several analyzed water samples contained cypermethrin at concentrations greater than 0.01 ppb, which means 76.2% or higher may cause mortality after 35 days. The farming shrimp activities in Tra Vinh cause soil

degradation, such as acidity, salinization, and spread of toxic alum substances (Al^{3+} , Fe^{2+} , H^+) from the soil into water (especially in Long Khanh, Giong Trom, Cai Doi, Dinh Cuu). Organic chemicals contaminate water in most canals, NH_3 , COD, chromaticity, and SS were higher than the allowed standard, so timely measures are required to resolve environmental pollution from farming shrimp activities in Tra Vinh.

Conflict of interest. The authors declare no conflict of interest.

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Authors:

Luom Thanh Thai, University of Kien Giang, Faculty of Natural Resource and Environment, Kien Giang 920000, Vietnam, e-mail: thaithanhluom@gmail.com

Huy Ba Le, Ho Chi Minh City University of Food Industry, Ho Chi Minh City 700000, Viet Nam, e-mail: lhuyba@gmail.com

Quang Minh Dinh, Can Tho University, School of Education, Department of Biology, Can Tho 900000, Vietnam, e-mail: dmquang@ctu.edu.vn

Hoan Xuan Nguyen, Ho Chi Minh City University of Food Industry, Ho Chi Minh City 700000, Viet Nam, e-mail: hoannx@hufi.edu.vn

Hiep Tan Dang, Ho Chi Minh City University of Food Industry, Ho Chi Minh City 700000, Viet Nam, e-mail: hiepdtd@hufi.edu.vn

Thanh Minh Le, Ho Chi Minh City University of Food Industry, Ho Chi Minh City 700000, Viet Nam, e-mail: thanhlek7@gmail.com

Ngoc Thi Hong Dang, University of Kien Giang, Faculty of Natural Resource and Environment, Kien Giang 920000, Vietnam, e-mail: dthngoc@vnkgu.edu.vn

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