Paddy Water Environ (2010) 8:361–369
DOI 10.1007/s10333-010-0215-5

ARTICLE

Pesticide discharge and water management in a paddy catchment in Japan

Thai Khanh Phong · Katsunori Yoshino · Kazuaki Hiramatsu · Masayoshi Harada · Tsuyoshi Inoue

Received: 1 February 2010 / Revised: 5 July 2010 / Accepted: 12 July 2010 / Published online: 22 July 2010

Abstract Concentrations of several pesticides were monitored in a paddy block and in the Kose river, which drains a paddy catchment in Fukuoka prefecture, Japan. Detailed water management in the block was also monitored to evaluate its effect on the pesticide contamination. The concentrations of applied pesticides in both block irrigation channel and drainage canal increased to tens of μg/L shortly after their applications. The increase in pesticide concentrations was well correlated with the open of irrigation and drainage gates in the pesticide-applied paddy plots only 1–3 days after pesticide application. High concentration of other pesticides, mainly herbicides, was also observed in the inflow irrigation and drainage waters, confirming the popularity of early irrigation and drainage after pesticide application in the area. The requirement of holding water after pesticide application (as a best management practice) issued by the authority was thus not properly followed. In a larger scale of the paddy catchment, the concentration of pesticides also increased significantly to several μg/L in the water of the Kose river shortly after the start of the pesticide application period either in downstream or mid–upstream areas, confirming the effect of current water management to the water quality. More extension and enforcement on water management should be done in order to control pesticide pollution from rice cultivation in Japan.

Keywords Rice pesticide · Paddy block · Drainage canal · Irrigation channel · Pesticide concentration

T. K. Phong (✉) · K. Hiramatsu · M. Harada
Department of Agro-Environmental Sciences, Faculty of Agriculture, Kyushu University, Fukuoka 812-8581, Japan
e-mail: thaikhanhphong@yahoo.com

K. Yoshino
Department of Bioproduction Environmental Sciences, Graduate School of Bioresource and Bioenvironmental Sciences, Kyushu University, Fukuoka 812-8581, Japan

T. Inoue
Water Supply Authority of Southern Fukuoka Prefecture, Fukuoka 830-0062, Japan

Introduction

Nonpoint source pollution due to agricultural chemicals including pesticide has been regarded as a threat to the quality of river waters, which constitutes the primary source of drinking water for many regions. Accounting for 50% of Japan's agricultural lands, rice paddy fields are thought to contribute significantly to the contamination of Japanese rivers by rice pesticides. Monitoring studies on river systems in Japan have detected several pesticides commonly used in paddy fields and the maximum concentrations ranged up to few tens of μg/L (Tanabe et al. 2001; Sudo et al. 2002, 2005; Nakano et al. 2004; Vu et al. 2006).

Water management is known as a key point in preventing pesticide discharge from paddy fields to the environment (Watanabe et al. 2008). In the past, Japanese farmers often practiced the spill-over water management that released considerable amount of pesticides to the drainage canals and then to the rivers. But with the increase in public awareness about the agricultural pollution, farmers are recommended to hold the water inside their paddies for several days after pesticide application. The

Japanese authority has recently increased the recommended water holding period after pesticide application from 3–4 to 7 days to improve its effectiveness. However, the above mentioned measure is not mandatory and there is no punishment for not following the recommendation. Some farmers, due to time constraint or lack of awareness and responsibility, still release lot of water from their plots to the drainage canals shortly after pesticide application. Previous studies (Sudo et al. 2002; Nakano et al. 2004; Kawakami et al. 2006), while reporting the pesticide contamination, did not provide detailed information on actual water management in the field, especially the period during and after pesticide application. Tanabe et al. (2001) and Nakano et al. (2004) reported the relation between pesticide concentration in the river and the pesticide application period but not the water management in paddy plots. Meanwhile, Vu et al. (2006) reported a general distribution of water level in plots of a small paddy catchment in a single day but failed to mention its closeness to the pesticide application period.

This study aims to fill the gap by monitoring the water management in paddy plots as well as the contamination of canal and river water due to the discharge of pesticide from paddy fields during the rice crop season. Then a relation between them can be derived for the purpose of planning a better strategy to control pesticide contamination from paddy fields.

Materials and methods

Study area

This study was carried out in the Kose river, which drains a paddy catchment of about 20 km^2 in Kurume city, Fukuoka prefecture, Japan. The Kose river originates from a reservoir and flows through agricultural lands before joining the Chikugo river.

Along its course, the Kose river (10–20 m wide) receives water from small springs, tail of irrigation and drainage water of farm lands located at the hillside of a mountain range. Those farm lands, most of them rice paddies and vineyard, are irrigated by water from upstream reservoirs. The monitored catchment and a paddy block subjected to intensive monitoring were shown in Fig. 1.

The monitoring was conducted in June and July 2009 during the rice crop season in this region. Water samples were taken from three sampling points in the Kose river, relatively located upstream, middle stream and downstream of the catchment. In the intensive monitoring block, water samples were taken from the entrance and outlet of both irrigation channel and drainage canal. Sampling points were also shown in Fig. 1.

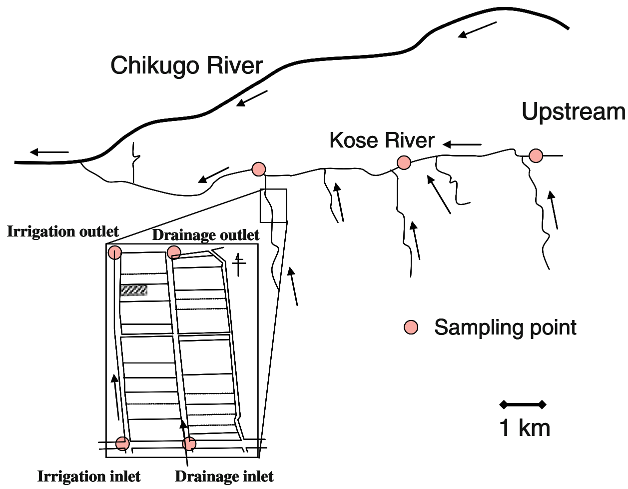


Fig. 1 Layout of the studied area

Intensive monitoring in a paddy block

An intensive monitoring was carried out in a 10-ha paddy block located at the downstream section of the Kose river to investigate the relation between pesticide contamination and water management. The layout of the block was shown in Fig. 1. The actual number of cultivating plots was 15 with individual plot size ranging from 0.1 to 1.1 ha. The block was consolidated recently with concrete bunds, new irrigation system and sub-surface drainage system. Irrigation channels are separated from the drainage canal but sometime can receive drainage water in emergency cases. A large drainage canal (2 m wide) flows in the middle of the block so that it could receive drainage water from plots lying in its 2 sides. The drainage canal initiates from upstream area and contains the drainage water of other paddy plots in the upstream area.

At the beginning of the study, questionnaires were sent to farmers (15 people) to collect information about their farming schedule and the possible use/application of pesticides. Information on pesticide application was compiled and later used for comparison with monitoring data. Water management in each plot including irrigation and drainage practices, was monitored daily around the pesticide application period and then twice a week since 2 weeks after pesticide application period. In order to verify the water management, water level and other water balance components including precipitation, evapotranspiration and percolation rate (Fig. 2) were also monitored during the monitoring period.

Velocity and depth of water flows in irrigation canal and drainage canal were also monitored. The data were then used to calculate the water flow in such canals. The difference in inflow and outflow would again confirm the occurrence of irrigation and drainage in the block. Because

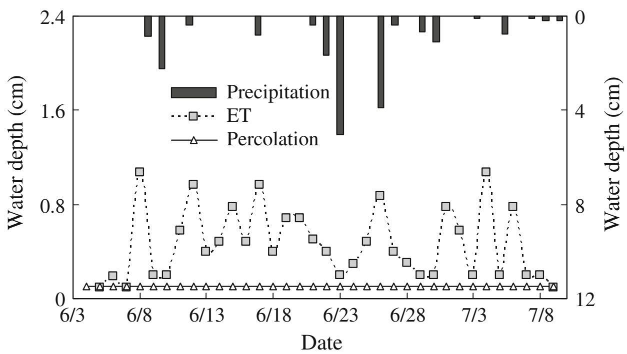


Fig. 2 Daily precipitation, evaporation (ET) and percolation in the block during the monitoring period

both irrigation and drainage canals were made of concrete and the average retention time was less than 30 min the adsorption and degradation of pesticides in the canals could be neglected.

Water sampling and chemical analysis

At each sampling point, water samples of 1-L were taken. Each sample was composited from several sub-samples taken from several spots at the site. After sampling, the samples were immediately transported back to the laboratory and were filtered before they were stored at 4°C prior to analysis. Samples were taken intensively during the pesticide application period of the paddy block (early June) and twice a week during the later part of the monitoring period.

An established method was used to analyze a range of pesticides in the water samples including 11 fungicides, 20 herbicides and 11 insecticides (with 4 metabolites). The list of monitored pesticides was presented in Table 1. This list includes some of the most popular pesticides used in rice cultivation in Japan.

Water samples were filtered again before extraction and 500 mL of water was solid phase extracted using a GL Science Aqusis PLS-3 cartridge (GL Science, Tokyo, Japan). Prior to use, the cartridges were initially conditioned with 10 mL of dichloromethane, 5 mL of methanol, followed by 5 mL of distilled water. The water sample was then loaded into the cartridge at a flow rate of 15 mL/min using the automated solid phase extraction equipment AutoTrace® SPE Workstation (Caliper Life Science, MA, USA). The cartridges were dried for 10 min by nitrogen gas before the pesticides were eluted by 3 mL of dichloromethane. The dichloromethane extracts were collected and evaporated to approximately 0.4 mL using a TurboVap LV Evaporator (Zymark Corporation, MA, USA). The residual solution was made up to 0.5 mL using pure dichloromethane and then transferred to the vial for gas chromatography analysis. Before analysis, 2 μL of a mixture of internal standards (anthracene-d_{10}; acenaphthene-d_{10}; p-terphenyl-d_{14}) at the concentration of 50 mg/L was added to the vial.

The pesticides were analyzed using a TRACE GC Ultra gas chromatograph (Thermo Fisher Scientific, MA, USA) equipped with a TSQ Quantum triple quadrupole mass spectrometer (Thermo Fisher Scientific, MA, USA). The column was a SLB-5MS column (30 m × 0.25 μm × 0.25 mm) (Supelco, PA, USA). Helium was used as the carrier gas. The temperature was programmed as follows: 60°C for 1 min, ramped up to 150°C at 20°C/min, then to 280°C at 10°C/min. The temperature was then maintained at 280°C for 5 min. A splitless injection mode was used, with an injection volume of 1 μL with the injection port hold at 240°C. The carrier gas was maintained constant at 1.2 mL/min. The mass spectrometer was set in selective reaction monitoring (SRM) mode. The limits of detection of all pesticides were 0.01 μg/L. The recovery ratios ranged from 90 to 110%. The analysis was performed at the laboratory of the Water Supply Authority of Southern Fukuoka Prefecture, Japan.

Results and discussion

Detection of pesticide in water samples

Among the total of 46 monitored compounds, 14 compounds (12 pesticides and 2 metabolites) were not found in all water samples and 2 other compounds detected one or two times at the detection limit (Table 1). As the study spanned for 2 months, it is possible that those pesticides were not used in this region. Most of the undetected pesticides were fungicides and insecticides, suggesting that these pesticide categories were not frequently used in the area. Meanwhile, only 2 herbicides were not found (in total 20 herbicides monitored), confirming the fact that herbicides were still the most used pesticides in Japan rice cultivation for the purpose of laborsaving.

Pesticide concentration in the paddy block

About 20 pesticides were found in water of both irrigation channel and drainage canal of the monitored paddy block from the start of the monitoring although only 7 monitored pesticides (all herbicides) were declared used in the block during the monitoring (questionnaires from farmers). It indicated that pesticides discharged from paddy plots in upstream area were taken with irrigation water and drainage water into the block.

In the studied block, herbicides were applied about 1 week after rice transplanting, in early June. The

Table 1 Concentrations (in μg/L) and frequency of pesticides found in water samples of the present study

	Block irrigation				Block drainage				Kose river					
	Inlet		Outlet		Inlet		Outlet		Upstream		Midstream		Downstream	
	Max[b]	*n*	Max	*n*	Max	*n*	Max	*n*	Max	*n*	Max	*n*	Max	*n*
Fungicide														
EDDP	ND		ND		ND		ND		ND		ND		ND	
Fthalide	ND		ND		ND		ND		ND		ND		ND	
Flutolanil	0.01	8.3	0.01	11.8	0.01	41.2	0.02	58.8	0.04	12.5	0.02	18.8	0.02	10.0
IBP	ND		ND		ND		ND		0.15	31.3	0.15	43.8	0.10	45.0
Isoprothiolane	0.13	75.0	0.11	88.9	0.13	82.4	0.40	88.2	0.19	75.0	0.14	75.0	0.14	85.0
Mepronil	ND		ND		ND		ND		ND		ND		ND	
Metalaxyl	ND		ND		ND		ND		0.10	37.5	0.11	50.0	0.08	80.0
Pencycuron	ND		ND		ND		ND		ND		ND		ND	
Procymidone	ND		ND		ND		ND		ND		ND		ND	
Pyroquilon	0.05	66.7	0.03	53.5	0.05	64.7	0.11	58.8	0.08	43.8	0.06	43.8	0.05	50.0
Sylafluofen	ND		ND		ND		ND		ND		ND		ND	
Herbicide														
Alachlor	ND		ND		ND		ND		ND		ND		ND	
Benthiocarb	0.81	75.0	0.16	71.5	0.16	47.1	0.54	58.8	ND		ND		0.02	15.0
Bromobutide	0.62	83.3	0.75	77.1	0.25	70.6	0.18	70.6	1.01	87.5	1.32	87.5	0.80	90.0
Cafenstrole	16.95	75.0	17.46	71.5	3.75	52.9	5.93	58.8	0.20	43.8	0.14	56.3	0.25	80.0
Clomeprop	ND		ND		ND		ND		ND		0.02	25.0	0.04	15.0
Cyhalofop butyl	0.01	16.7	0.04	11.8	0.06	5.9	0.08	5.9	ND		ND		0.02	10.0
Debromobutide	ND		ND		ND		ND		0.02	12.5	0.01	12.5	0.01	30.0
Dimepiperate	ND		ND		ND		ND		ND		ND		ND	
Dimethametryn	0.79	83.3	1.70	100.0	1.59	58.8	5.69	88.2	0.31	81.3	0.14	81.3	0.42	85.0
Esprocarb	0.02	25.0	0.02	18.1	0.02	17.6	0.02	11.8	0.04	25.0	0.37	37.5	0.14	50.0
Mefenacet	9.42	83.3	9.85	100.0	13.10	88.2	10.94	100.0	0.94	87.5	1.35	87.5	5.31	90.0
Molinate	0.04	8.3	ND		0.01	5.9	ND		0.01	6.3	ND		0.01	5.0
Oxaziclomefone	0.02	8.3	0.10	22.2	0.02	5.9	0.19	52.9	0.01	6.3	0.02	25.0	0.02	20.0
Pretilachlor	5.13	83.3	7.00	94.4	8.96	70.6	15.89	82.4	1.95	81.3	0.84	81.3	2.70	80.0
Pyributicarb	ND		ND		ND		0.32	11.8	0.02	6.3	0.04	18.8	0.07	30.0
Pyriftalid	0.02	8.3	0.02	11.8	0.04	5.9	0.03	5.9	0.01	6.3	0.01	18.8	0.02	15.0
Pyriminobac methyl	0.40	16.7	0.99	34.0	0.40	17.6	1.69	52.9	0.02	18.8	0.06	37.5	0.11	45.0
Simazine	ND		0.01	5.6	0.01	5.9	0.02	23.5	ND		ND		0.03	10.0
Simetryn	ND		ND		ND		ND		ND		ND		ND	
Thenylchlor	ND		0.02	11.8	0.02	5.9	0.51	23.5	0.03	18.8	0.09	25.0	0.21	50.0
Insecticide														
BPMC	ND		ND		ND		ND		ND		ND		ND	
Buprofezin	0.15	91.7	0.06	82.6	0.12	70.6	0.11	76.5	0.09	81.3	0.08	75.0	0.10	70.0
DDVP	0.05	16.7	0.03	6.3	0.02	11.8	0.02	17.6	0.03	25.0	0.15	25.0	0.08	55.0
Diazinon	0.03	25.0	0.03	24.3	0.44	41.2	0.61	47.1	ND		0.03	6.3	0.03	25.0
Diazoxon	ND		ND		ND		ND		ND		ND		ND	
DMTP[a]	0.39	75.0	0.25	70.1	0.16	70.6	0.15	64.7	0.12	62.5	0.13	50.0	0.16	90.0
Etofenprox	ND		ND		ND		ND		ND		ND		ND	
Fenthion	ND		ND		ND		ND		ND		ND		ND	
Fenthion sulfon[a]	ND		ND		ND		ND		ND		ND		ND	
Fenthion sulfoxide[a]	ND		ND		ND		ND		ND		ND		ND	
Isoprocarb	ND		ND		ND		ND		0.02	6.3	0.01	6.3	0.02	5.0
Malathion	ND		ND		ND		ND		ND		0.01	6.3	0.01	5.0
MEP	0.18	58.3	0.03	35.4	0.16	52.9	0.14	35.3	0.03	18.8	0.04	31.3	0.10	40.0
MEP oxon[a]	0.04	8.3	0.02	6.3	0.02	5.9	0.02	5.9	ND		ND		ND	
Pyridaphenthion	ND		0.01	5.6	0.02	5.9	ND		ND		ND		ND	

n = (number of samples containing pesticide/number of samples analyzed) × 100

[a] Metabolite

[b] Maximum concentration detected

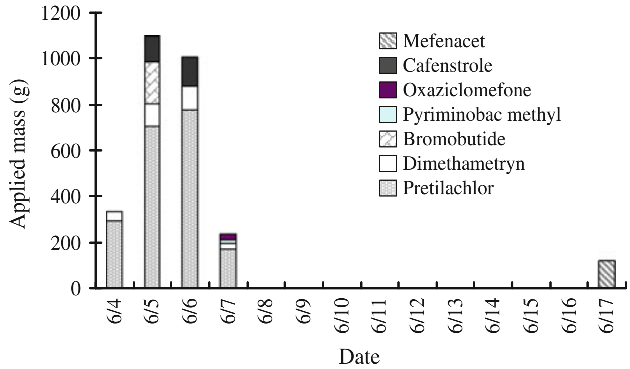
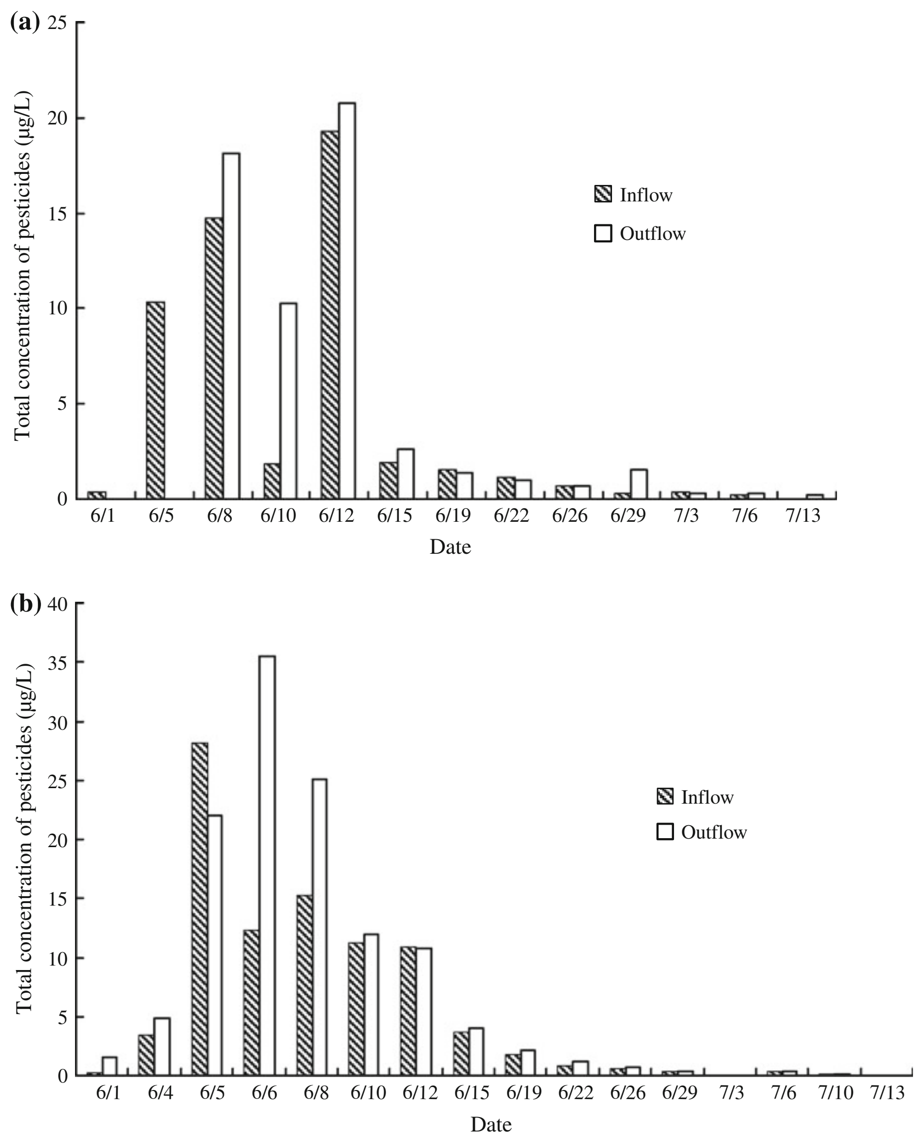


Fig. 3 Mass distribution of applied pesticides in the studied block

distribution of pesticide application in the block was shown in Fig. 3 (for 7 monitored pesticides). The application period ended within 4 days except in one tiny plot with late transplantation. Most of applications were carried out in 2 week-end days (June 6 and 7) as many farmers are part-time farmers who have another job during weekdays. The surrounding downstream area also had the same cropping time and practice. The main active ingredient used in the block was pretilachlor, which was accompanied by dimethametryn. Other popular compounds included cafenstrole and bromobutide.

High pesticide concentrations were found in irrigation water and drainage water of the block during the pesticide application period. Total concentration of pesticides in those waters during the monitoring period was shown in Fig. 4. The total concentration of pesticides has been used as an indicator of water quality and the European Union has set a limit of 0.5 μg/L for total concentration of pesticides in water used for drinking purpose (98/83/EC). Nevertheless, in the period close to the pesticide

Fig. 4 Total concentration of pesticides in **a** irrigation and **b** drainage canals



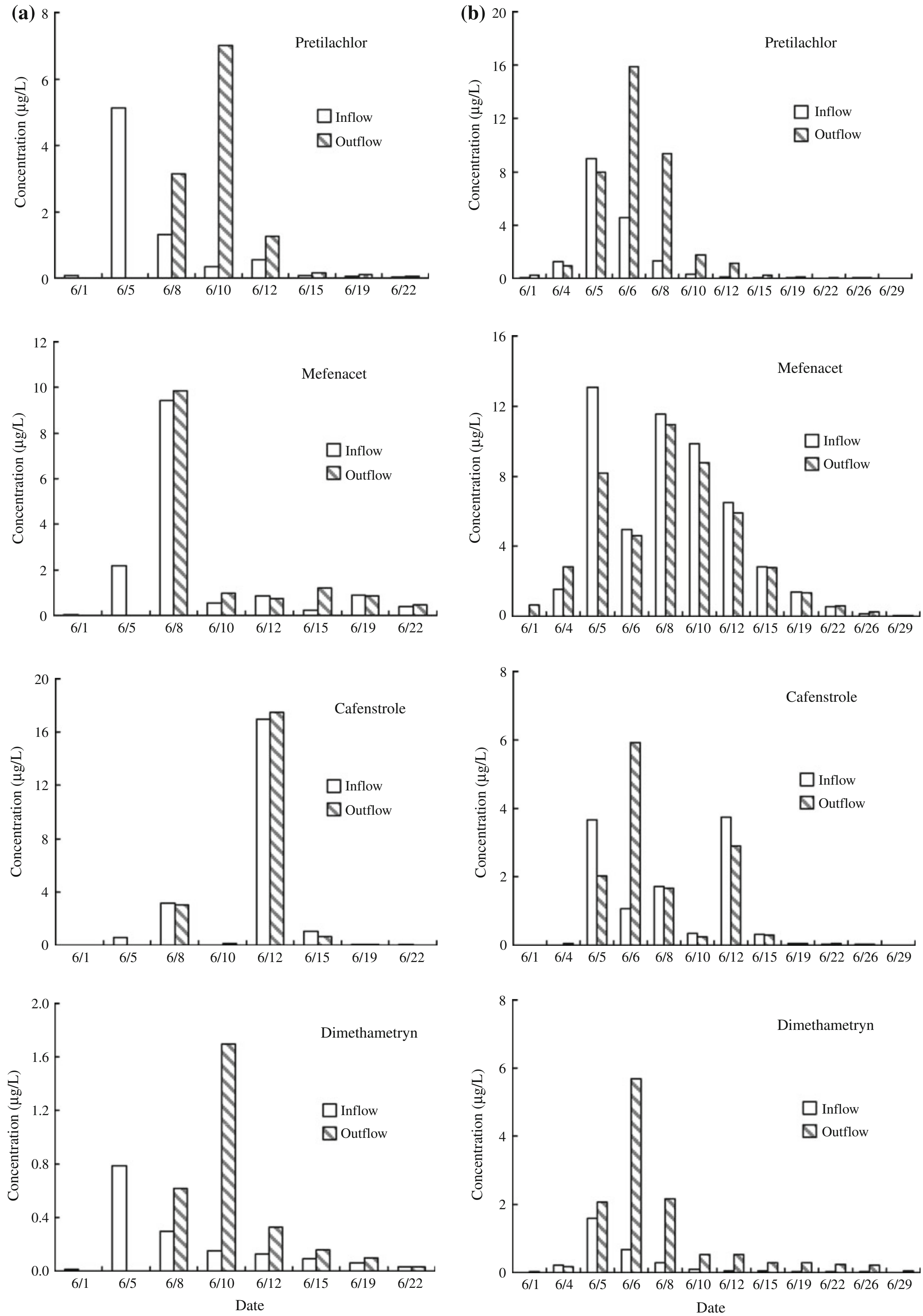


Fig. 5 Concentration of major pesticides in **a** irrigation and **b** drainage water

application the total concentration of pesticides in the samples of the block was far above this limit. Maximum values were 20.8 µg/L in outflow irrigation water and 35.5 µg/L in outflow drainage water, respectively. From the end of June, which was about 3 weeks after pesticide application period, the total concentration of pesticides fell below the 0.5 µg/L threshold.

Not every pesticide contributed equally to the sum of individual pesticide concentrations. While insecticide and fungicide concentrations remained at residual level of about 0.1 µg/L or less, concentrations of some major herbicides reached up several µg/L around the pesticide application period (Fig. 5). The maximum concentration in irrigation water was that of cafenstrole (17.5 µg/L) and the maximum concentration in drainage water was that of pretilachlor (15.9 µg/L). Concentrations of major herbicides in the drainage canal were comparable to those reported by Vu et al. (2006) in a drainage canal of a paddy block in Ibaraki prefecture, Japan. Meanwhile, Nakano et al. (2004) recorded concentration of up to 90 µg/L at the outlet of a paddy block's drainage canal, where farmers may practice spillover irrigation.

It is noted that the irrigation water of the studied block was also contaminated with pesticides. The cause of this phenomenon is due to the structure of the irrigation gate in each paddy plot (Fig. 6). With this structure, water can either enter or drain from the plot depending on the difference in water level between the irrigation channel and the paddy plot. This irrigation gate can sometime be used intendedly to drain water quickly in case of emergency (for example, draining water to treat the snail problem).

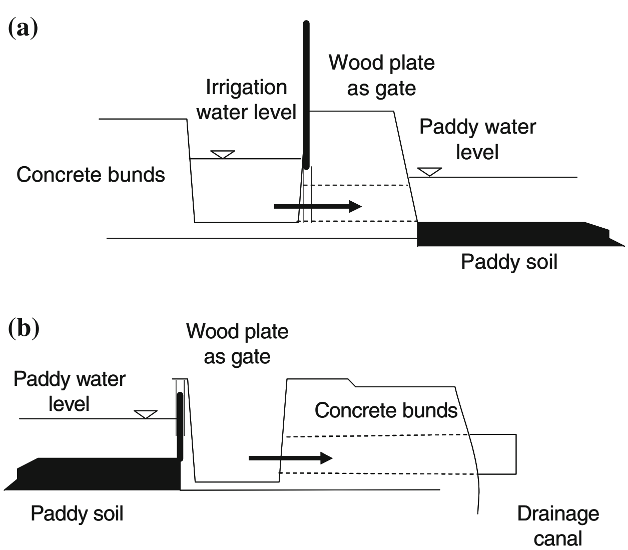


Fig. 6 Design of irrigation (**a**) and drainage (**b**) gates in the studied paddy block

Effect of water management on the pesticide concentration in the paddy block

Observation of Figs. 2, 3, and 4 indicated that pesticides were discharged from paddy plots to the environment shortly after pesticide application. This was because the water was not managed appropriately in the studied block as well as in the surrounding area.

The application of water holding period (7 days as recommended in the pesticide labels and required by Japan authorities), during which the drainage was completely stopped, was supposed to be followed in order to reduce the load of pesticide discharge. Previous studies have proved the effectiveness of this water management in controlling pesticide discharge from paddy fields (Vu et al. 2006; Watanabe et al. 2007). The farmers were also aware of this requirement as indicated by their answers in the questionnaires. However, the actual water management in the studied block was not satisfactory (Fig. 7).

Most of the plots drained only 1 or 2 days after pesticide application (Fig. 7a). Because the pesticide concentration in paddy plot water is highest at that moment, the drained

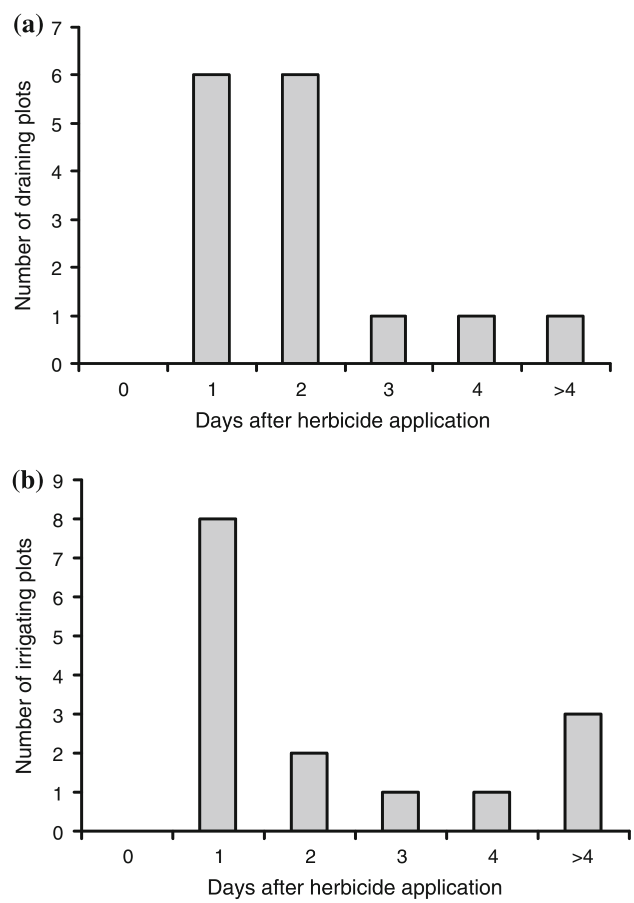
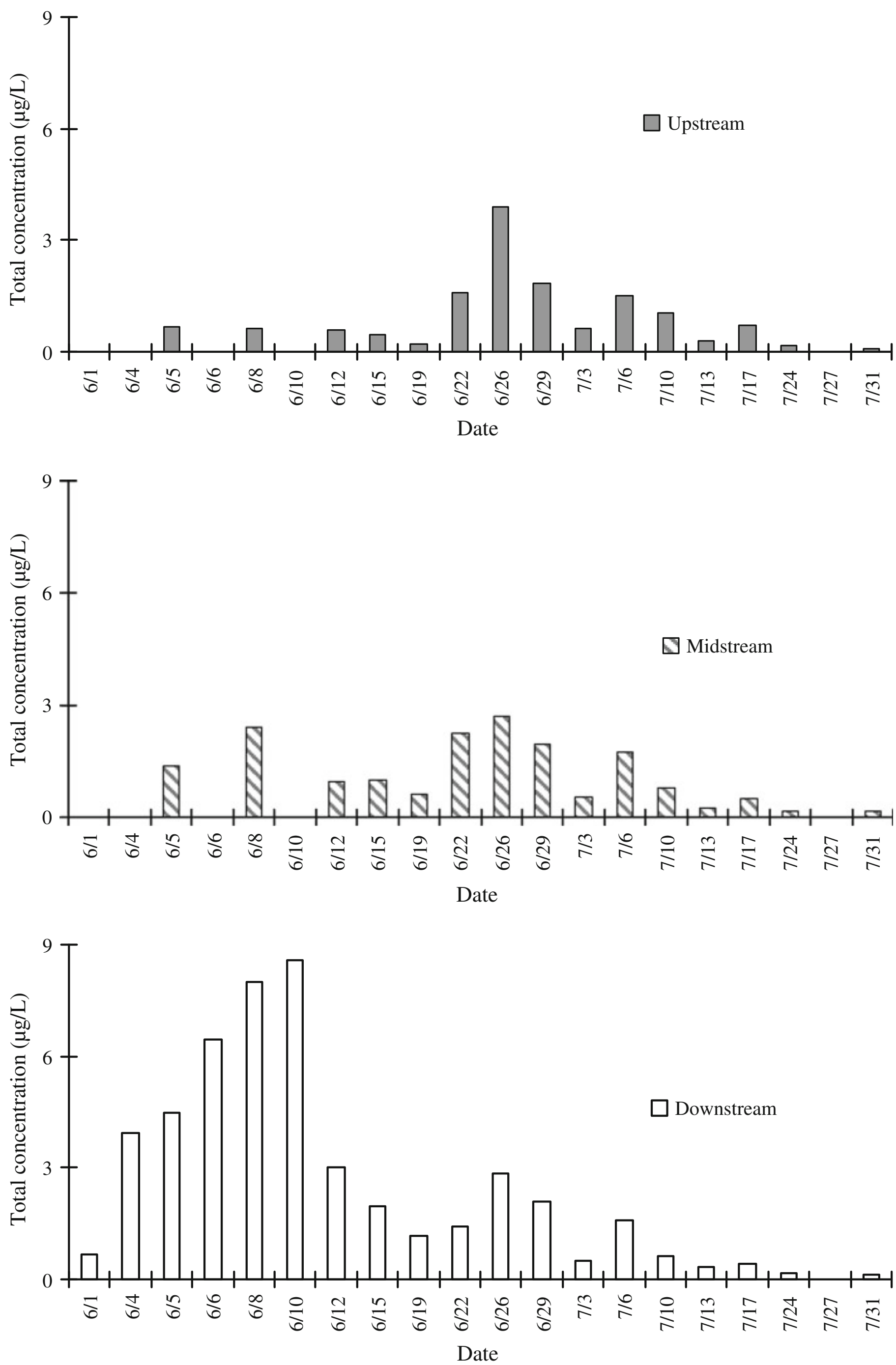


Fig. 7 The first draining (**a**) and irrigating (**b**) events in paddy plots of the monitored block after their pesticide application

water would also contain very high concentration of applied pesticides. Therefore, the early drainage practice eventually led to the discharge of remarkable amount of pesticides into the irrigation canal and the drainage canal. It was clearly demonstrated by the significant difference between inflow and outflow concentration of pretilachlor, cafenstrole and dimethametryn (Fig. 5). It was confirmed that the plots applied with pretilachlor, cafenstrole and dimethametryn were among those draining only 1 or 2 days after pesticide application. Their outflow concentrations steeply increased from the inflow concentrations only 2 days or even 1 day after their respective application. For the case of mefenacet, its concentration remained similar or became lower in the outflow water due to the dilution effect as mefenacet was not applied until June 17 in a small amount (Fig. 5).

Irrigation to treated plots was also initiated only 1–3 days after pesticide application (Fig. 7b). As mentioned above,

Fig. 8 Total concentrations of pesticides in the Kose river



the structure of the irrigation gate in this area allowed paddy water to be released into irrigation channel when the gate is open. And similar to the phenomenon in the drainage canal, the concentrations of pretilachlor and dimethametryn increase in outflow irrigation water shortly after their application to the plots.

Pesticide concentration in the river

Concentration of pesticides in the Kose river was lower than that in the block drainage canal (Fig. 8). The highest total concentration of pesticides occurred in downstream section of the river, reaching 8.6 μg/L during the pesticide application period of the downstream area. Similar to the case of the drainage canal, concentrations of some major herbicides accounted for most of the summed value. Concentrations of pretilachlor and mefenacet in the Kose river (data not shown) were comparable to those recorded previously in other rivers in Japan (Nakano et al. 2004; Sudo et al. 2005).

Due to the lag in rice transplanting time between downstream area and midstream–upstream area, the peak of pesticide concentration occurred differently between those areas (Fig. 8). As the transplanting time of downstream area was around end of May, early June and that of midstream–upstream area was end of June, it was observed that the concentration of pesticides in the Kose river increased shortly after the start of the pesticide application period. Probably, water management for the purpose of preventing pesticide pollution may still not be well practiced in some regions.

Conclusions

Concentrations of several pesticides were monitored in a paddy block and in the Kose river in Fukuoka Prefecture, Japan. In both block irrigation channel and drainage canal, the concentrations of applied pesticides increased shortly after their application. Those augmentations in pesticide concentrations were well correlated with the open of irrigation and drainage gates in the pesticide-applied paddy plots. The requirement of holding water after pesticide application (as a best management practice) was not properly followed in most of the cases. Similar situation happened in the catchment scale when the concentration of pesticides also increased in the water of Kose river shortly after the start of the pesticide application period either in downstream or mid–upstream areas. More attention should be paid to agricultural extension and enforcement in order to reduce pesticide pollution from rice cultivation in Japan.

Acknowledgments Thanks are due to the officers and farmers in the Takeno paddy block for their cooperation. This paper was produced when TK Phong is a JSPS postdoctoral fellow at Faculty of Agriculture, Kyushu University. This research is partly supported by the JSPS Grant-in-Aid for Scientific Research (Project number: P08439).

References

98/83/EC. The Council Directive of 3 November 1998 on the quality of water intended for human consumption

Kawakami T, Eun H, Arao T, Endo S, Ueji M, Tamura K, Higashi T (2006) Concentration and loading of several pesticides in water, suspended solids and sediment during ordinary water discharge in Sugao marsh, Ibaraki Prefecture, Japan. J Pest Sci 31(1):6–13

Nakano Y, Miyazaki A, Yoshida T, Ono K, Inoue T (2004) A study on pesticide runoff from paddy fields to a river in rural region-1: field survey of pesticide runoff in the Kozakura River, Japan. Water Res 38(13):3017–3022

Sudo M, Kunimatsu T, Okubo T (2002) Concentration and loading of pesticide residues in Lake Biwa basin (Japan). Water Res 36(1):315–329

Sudo M, Okubo T, Kaneki R (2005) Paddy herbicide inputs in the entire river inflow reaching Lake Biwa, Japan. Limnology 6(2): 91–99

Tanabe A, Mitobe H, Kawata K, Yasuhara A, Shibamoto T (2001) Seasonal and spatial studies on pesticide residues in surface waters of the Shinano river in Japan. J Agric Food Chem 49(8):3847–3852

Vu SH, Ishihara S, Watanabe H (2006) Exposure risk assessment and evaluation of the best management practice for controlling pesticide runoff from paddy fields. Part 1: Paddy watershed monitoring. Pest Manag Sci 62(12):1193–1206

Watanabe H, Nguyen MHT, Souphasay K, Vu SH, Phong TK, Tournebize J, Ishihara S (2007) Effect of water management practice on pesticide behavior in paddy water. Agric Water Manage 88(1–3):132–140

Watanabe H, Inao K, Vu HS, Phong TK, Ishihara S, Takagi K, Tournebize J (2008) Pesticide exposure assessment in rice paddy areas: a Japanese perspective. In: Capri E, Karpouza D (eds) Pesticide risk assessment in rice paddies: theory and practice. Elsevier, Amsterdam, pp 167–211