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Exposure risk assessment and evaluation of the best management practice for controlling pesticide runoff from paddy fields. Part 1: Paddy watershed monitoring

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Abstract: Rice pesticide concentrations in surface water along with hydrological balance and water management conditions were investigated in a paddy watershed of about 100 ha at the Sakura river basin in Tsukuba, Japan, for 3 years from April 2002. Monitoring on different hydrological scales ranging from a paddy plot up to a watershed determined the importance of water management associated with rainfall events and the cyclic irrigation for reducing pesticide discharge into aquatic environments. Surface drainage significantly increased as a response to rainfall events greater than about 1.5 cm day^{-1}. A total of 16 herbicides were detected in the stream water and their peak concentrations mostly occurred from early to mid-May following the pesticide application period. Two water management factors influencing the pesticide runoff from paddy fields were defined: excess water storage capacity (EWSC) and water holding period (WHP). Uncertainty analyses of pesticide discharge from a paddy plot for dymron (daimuron) and imazosulfuron (IMS) were performed using Monte Carlo simulation (MCS) with prescribed probability of rainfall and water management practice from observations over a period of 3 years. Application of an intermittent irrigation scheme with shallow water depth practice and high drainage gate to maintain the EWSC >2 cm and increasing WHP from the current Japanese Agricultural Chemicals Regulation law of 3–4 days to at least 10 days were recommended for reducing the pesticide runoff from paddy fields in a monsoon region such as in Japan. The combination of good water management in field plots and small-scale water cycling is the best management practice for controlling pesticide discharge from paddy watersheds.

Keywords: rice paddy field; pesticide; monitoring; watershed; best management practice; uncertainty; Monte Carlo

1 INTRODUCTION

About 98% of all of the pesticides sold in Japan are used in agriculture and forestry, of which 50% are used for paddy rice production.[1] Improper field water management and large precipitation events may result in appreciable paddy runoff and pesticide discharge from rice production. Peak pesticide concentrations in nearby surface water bodies may exceed toxic levels for aquatic organisms and be unacceptable with regard to drinking water standards. Pesticide monitoring studies in Japanese river systems have found concentrations of commonly used rice pesticides of up to 10 μg L^{-1},[1–5] which have caused adverse effects on the aquatic ecosystem.[3,6,7] Public concern regarding the impact of rice pesticides on surface water quality is increasing.

For the protection of the aquatic ecosystem and human health from pesticide pollution, it is important that rice growers practise good water management in relation to meteorological conditions to achieve water quality goals.[8,9] Concerning water management, the water holding period requirement in rice fields after application is one of the main factors that significantly reduces pesticide loadings in the receiving water.[9,10]

Attempts have been made to investigate a more favourable water management programme for the reduction of pesticide discharge from rice fields into the aquatic environment; however, these have so far mainly concentrated on the field plot scale. Based on the monitoring of pesticide dissipation in paddy plots, it was suggested that water holding times be increased from the Japanese current recommendation of 3–4 days to 1 week or more.[9,11] Watanabe *et al.*[12] examined the pesticide dissipation in rice paddies using the pesticide fate model PCPF-1 with different water management scenarios, and recommended minimum surface drainage and optimum ponding water depth to reduce pesticide losses. Along with the watershed level, hydrological processes including water management practices in rice fields are probably the main factors affecting pesticide loading in the aquatic environment. However, studies dealing with the fate and transport of rice pesticides on a watershed

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scale are limited and mainly focused on the calculation of total pesticide losses[4,5,13] and the calibration of the pesticide fate models.[5,14,15] Thus, the optimum watershed management for improving surface water quality associated with rice pesticide pollution ought to be investigated.

The objectives of this study are (1) to monitor rice pesticide concentrations in surface waters along with hydrological conditions in a paddy watershed, and (2) to examine key factors affecting the pesticide runoff from rice fields for evaluation of the best management practice (BMP) for the reduction of pesticide discharge from paddy fields into the aquatic environment.

2 MATERIALS AND METHODS

2.1 Watershed description

The study area was a paddy watershed (Fig. 1) located in the Sakura river basin of Ibaragi prefecture, about 50 km north-east of Tokyo, Japan. The area of the watershed was about 97 ha, mainly covered by 86 ha of paddy fields. The watershed was reformed as standard paddy fields by a land consolidation project of the Japanese Ministry of Agriculture, Forestry and Fisheries. Paddy plots are similarly distributed on both sides of the drainage canals and are grouped into farm blocks, which are surrounded by farm roads and drainage canals. The average slope of the topography of the watershed is about 0.2%. The area of the paddy plots ranges from 0.2 to 0.4 ha, and their short sides face a farm drainage canal and an irrigation pipeline established along a farm road. Irrigation water is supplied to individual plots through pipelines from pump stations. The surface water for most of the paddy plots is gravitationally drained through the drainpipe to the drainage canal. Water supplied to the watershed consists of springs from Mt Tsukuba, which flow into the watershed through S1 and S4, external water from Lake Kasumigaura through a pipeline, and pumped stream water from the Sakasa River from P3 (Fig. 1). A part of the drainage water in the canals is reused for irrigation by pumping stations P1 and P2. The canals have a rectangular cross-section with concrete banks on both sides. The width of the canal ranges from 0.5 m upstream to 6 m at the outlet of the watershed (Table 1).

All the paddy plots within the watershed are cultivated with a single crop. Land preparation started from mid-April. Rice seedlings at the fourth leaf expanding stage are transplanted by transplanting machines in almost all the paddy fields in early May. For weed control, herbicides are often applied during a period of about 1–2 weeks after transplanting, depending on the herbicide product.[16] The water holding requirement of 3–4 days after application addressed in the pesticide labels is now a legal requirement in the new Agricultural Chemicals Regulation law that was revised in 2005 in Japan.[17]

2.2 Watershed monitoring

Monitoring of the hydrological balances was conducted for a paddy plot, farm block and watershed scales. Detailed descriptions of the monitoring stations in the watershed are shown in Fig. 1 and Table 1. A paddy plot of 0.3 ha (plot 1) and two farm blocks of 4.8 ha (block 1) and 5.3 ha (block 2) were selected as representatives of the hydrological response of the paddy plots and paddy blocks in the watershed. The monitoring period lasted 46 days from rice transplanting (1 May) to 15 June.

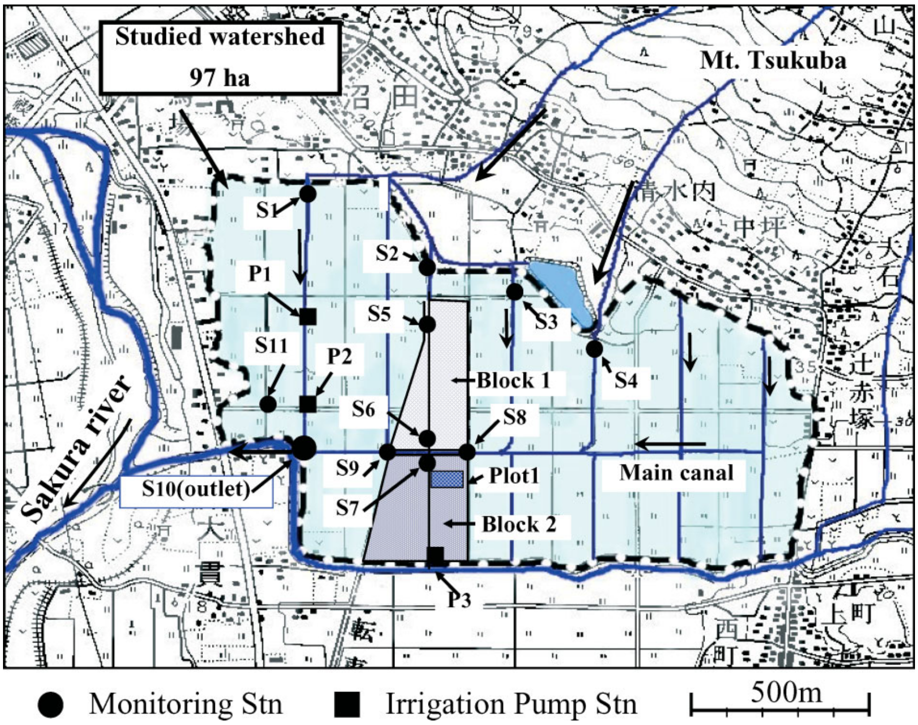

Figure 1. Studied watershed and monitoring stations.

Table 1. Monitoring stations

Stations	Location	Width of canal (m)	Paddy area[a] (ha)	Contents	Year
Paddy plot 1	Farm block 2	–	0.3	Irrigation, drainage water depth, water sampling	2002
S1, S2, S3, S4(inflow of watershed)	Secondary canal	0.5–3	–	Discharge	2003
S5(inlet of farm block 1)	Secondary canal	0.75	2.35	Discharge, water sampling	2002
S6(outlet of farm block 1)	Secondary canal	0.75	8.56	Discharge, water sampling	2002
S7(outlet of farm block 2)	Secondary canal	0.5	5.32	Discharge, water sampling	2002–2004
S8	Main canal	5	40.05	Discharge, water sampling	2002
S9	Main canal	5	54.12	Discharge, water sampling	2002
S10(outlet of watershed)	Main canal	6	86.1	Discharge, water sampling	2002–2004
S11(rain gauge)		–	–	Precipitation	2002–2004

[a] Total area of paddy fields covered from upstream to the monitoring station.

Precipitation was monitored by a rain gauge (No. 34-T; Ota keiki Co., Ltd, Tokyo, Japan) at site S11 of the watershed (Fig. 1). Other meteorological data including temperature, wind speed, humidity and solar radiation were obtained from a local meteorological station at Shimotsuma located about 5 km west of the study site.

For water management practices in paddy plot 1 in 2002, irrigation was monitored using a water tank attached to a V-notch weir and a water level sensor (HM–910; HI-NET Co., Ltd, Tokyo, Japan) with a data logger. Ponding water depth in the plots was also monitored by a water level sensor (HM–910; HI-NET Co., Ltd, Tokyo, Japan) having a data logger. Paddy runoff over the drainage gate with a flat weir was estimated from the paddy water level measured by the water level sensor. Daily evapotranspiration in the paddy fields was estimated by the FAO Penman–Monteith method[18] using meteorological data obtained from the meteorological station of Shimotsuma with a calibrated crop coefficient for Japanese rice.[19] The total vertical percolation and lateral seepage were computed from the water balance equation in paddy plot 1 [Eqn (1)] using the measured rainfall, irrigation, drainage and paddy water depth (the vertical percolation and lateral seepage were assumed to be the same for the entire watershed during the monitoring period):

$$
\begin{aligned}
\mathrm{d}H_\mathrm{w}/\mathrm{d}t = {} & \mathit{RAIN} + \mathit{IRR} - \mathit{DR} \\
& - \mathit{PERC} - \mathit{LSEEP} - \mathit{ET}
\end{aligned} \qquad (1)
$$

where H_w is the paddy water depth (cm), t is time (days), RAIN is the rainfall (cm day^{-1}), IRR is the irrigation (cm day^{-1}), DR is the surface drainage including the paddy runoff (cm day^{-1}), PERC is the vertical percolation (cm day^{-1}), LSEEP is the lateral seepage (cm day^{-1}) and ET is the evapotranspiration (cm day^{-1}).

Direct measurement of the surface discharge through drainpipes from the paddy plots was conducted about once a week for blocks 1 and 2. Rainfall data, surface discharge from the paddy plots and monitored discharge at S5, S6 and S7 were used for estimation of the lateral seepage from the paddy fields into the canals using the water balance equation to the canal segments within blocks 1 and 2.

To measure the discharges in the canal network, ten monitoring stations were established: S1 to S7 in the secondary drainage canals with an open-channel structure, and S8 to S10 in the main canal, where concrete weirs were located. These weirs were constructed during the period of the watershed consolidation project for irrigation head works. Discharge at S1 to S7 was estimated by the water depth–discharge curves obtained by a 5 min interval of continuous monitoring of the water depth using water level sensors (HM–910; HI-NET Co., Ltd, Tokyo, Japan) and weekly measurements of the average flow velocity with a flowmeter (AEM1-D; Alec Electronics Co., Ltd, Tokyo, Japan). The discharges at S8 to S10 were calculated by monitoring the water depth on the top of each weir using the equations described by Rao and Muralidhar.[20] In 2003, the hydrological balance in the watershed was evaluated using the observed data of inflows (at S1 to S4), outflow (at S10) and estimated water management practices in the paddy compartment of 86 ha.

In 2004, field surveying was conducted using questionnaires from growers of 19 plots in farm block 2. Information on pesticide use, including active ingredients, application rate and time, as well as paddy area and field practice, was successfully collected from the growers. The time of application is highly dependent on farming conditions and was assumed to follow a normal distribution with an estimated mean and a standard deviation using the data observed from farm block 2.

2.3 Water sampling and chemical analysis

Water samples were periodically taken about once a week starting from 30 April for irrigation water, paddy water (2002) and stream water at each station in the drainage canals (2002, 2003 and 2004). On the sampling days, water samples (200 mL) were collected at 1–2 cm below the water surface from eight spots in plot 1 and mixed in amber glass bottles. The concentrations in the mixed water sample represented the average concentration in the paddy water in plot 1. At the same time, water samples (about 2000 mL) were also taken at the drainage gate of plot 1 to monitor the concentrations in the drainage water. Samples of the stream water (2000 mL) were taken from several spots at the monitoring stations and were mixed in amber glass bottles.

The herbicides in the water samples included oxaziclomefone, simetryn, esprocarb, dimethametryn, dimepiperate, pretilachlor, pyriminobac-methyl, pyributicarb, pentoxazone, mefenacet, cafenstrole, molinate and thiobencarb, which were analysed using a gas chromatograph (GC/FTD, SHIMAZU GC-17A for 2002 and GC-20A for 2003–2004), and bensulfuron-methyl, imazosulfuron, pyrazosulfuron-ethyl and dymron (diamuron), which were analysed using a liquid chromatographic-tandem mass spectrometer (LC/MS/MS). Immediately after sampling, the water samples were pretreated by filtering through Whatman GF/B-1.0 µm and GF/F-0.7 µm glass filters and adjusting the pH of the filtrate to 6.5 with phosphoric acid solution (10%) or hydrochloric acid (2M). About 20 mL of the filtered sample was kept frozen at −20 °C for the LC/MS/MS analysis while the rest of the sample was used for the GC analysis.

For GC analysis, after preconditioning with dichloromethane (5 mL), methanol (5 mL) and distilled water (10 mL), filtrate (1 L) was passed through solid-phase extraction cartridges (Sep-Park®, tC18 for 2002 and PS-2 for 2003–2004) at a rate of 10 mL min^{-1}. The extracts were then eluted with dichloromethane (ca 10 mL), and the elute was dried under reduced pressure and reconstituted with acetone (2 mL). The extracted samples were stored at 4 °C before analysis. The column used in the GC was a DB-5 (J&W) column (30 m × 0.25 µm × 0.32 mm). The temperature was programmed as follows: 60 °C (2 min) ramped up to 140 °C at 10 °C min^{-1}, then to 270 °C at 5 °C min^{-1}. The temperature was held at 270 °C for 4 min. A splitless injection mode was used with an injected volume of 4 µL. The carrier gas pressure was set at 40 kPa for 2 min, then increased to 64 kPa at 3 kPa min^{-1} and continued to ramp at 1.5 kPa min^{-1} to 103 kPa, which was maintained for 4 min. The herbicide was detected by a flame thermoionic detector (FTD). The recoveries, determination limit and coefficient of variation obtained with three replications of the pesticides are shown in Table 2.

Table 2. Recovery, limit of determination and coefficient of variation of the pesticide by GC analysis

Herbicide	Recovery ratio (%)	Limit of determination (µg L^{-1})	Coefficient of variation (%)
Oxaziclomefone	107.94	0.016	18.81
Molinate	85.77	0.05	3.86
Symetryn	88.98	0.024	2.07
Esprocarb	85.01	0.024	3.76
Thiobencarb	87.57	0.016	2.89
Dimethametryn	90.51	0.0083	2.54
Dimepiperate	91.21	0.016	2.45
Pretilachlor	93.73	0.033	2.39
Pyriminobac-methyl (*E*)	95.57	0.017	2.81
Pyributicarb	106.22	0.016	11.64
Pentoxazone	105.40	0.044	8.89
Mefenacet	88.49	0.039	5.91
Cafenstrole	106.18	0.024	2.70

For the LC/MS/MS analysis, 0.5 mL of the filtered sample and 0.5 mL of acetonitrile containing primisulfuron-methyl (40 ng mL^{-1}) as the internal standard were well mixed and centrifuged for 10 min at 20 600 × *g*. A clear supernatant was then injected directly for the analysis. HPLC was performed using a SHISEIDO NANOSPACE SI-2 system equipped with an OptiGuard mini guard column (Optimize Technologies, OptiGuard mini C18, 15 × 1.0 mm) and a Cadenza column (Intakt, CD-C18, 30 × 2.0mm). Elution was performed in isocratic mode with 10 mM acetic acid + acetonitrile (45 + 55 by volume) at a liquid flowrate of 0.1 mL min^{-1}. The injection volume was 5 µL. Mass spectrometry was performed with an Applied Biosystems API 3000TM LC/MS/MS system, using TurboIonSpray® ionisation in the positive mode. The limit of determination of the four pesticides analysed by LC/MS/MS was 0.08 µg L^{-1}. The above pesticide analysis was performed at the National Institute for Agro-Environmental Sciences (NIAES) in Tsukuba, Japan.

2.4 Data analysis

In actual rice production, farmers manage water in their fields mainly on the basis of ponding depth. They start and stop irrigation when water depths go respectively below and above certain levels based on their experience. Therefore, for water management evaluation, the values of maximum and minimum practised paddy water depth, $H_{\mathrm{w\,max}}$ and $H_{\mathrm{w\,min}}$, were used instead of the volume of irrigated water. When the ponding water depth is below the height of the drainage gate, H_{gate}, the depth from this water level to the top of the drainage gate can be used to store excess water on rainfall events. This depth is defined as the excess water storage depth (EWSD). The average EWSD during the growing season represents the capacity of the field to store excess water input such as rainfall, and is defined as the excess water storage capacity (EWSC). On day i, surface discharge (DR_i) occurs only when the total input (*IRR* and *RAIN*) minus the total output (*PERC, ET, LSEEP*) exceeds the EWSD as presented in Eqn (2), and the EWSC of the fields is calculated by Eqn (3):

$$DR_i = RAIN_i + IRR_i - PERC_i - LSEEP_i - ET_i - EWSD_i \tag{2}$$

$$\mathrm{EWSC} = \frac{1}{n}\sum_{i=1}^{n} EWSD_i = H_{\mathrm{gate}} - H_{\mathrm{w}} \tag{3}$$

where DR_i, $RAIN_i$, IRR_i, $PERC_i$, $LSEEP_i$, $LSEEP_i$, ET_i and $EWSD_i$ are the surface discharge, rainfall, irrigation, vertical percolation, lateral seepage, evapotranspiration and excess water storage depth at day i (cm) respectively, n is the length of the growing season (day), H_{gate} is the height of the drainage gate (cm) and H_{w} is the average practised paddy water depth during the growing season (cm).

Daily and cumulative pesticide discharge from a paddy plot expressed by the percentage of the applied mass can be calculated using the following equation:

$$DLoss_i = 10C_iDR_iA \tag{4}$$

$$CumLoss = 100\frac{\sum_{i=1}^{n} DLoss_i}{A_{pp}A} \tag{5}$$

where $DLoss_i$ is the pesticide loss on day i (mg), C_i is the pesticide concentration in the paddy water on day i (mg L^{-1}), DR_i is the surface runoff or drainage on day i (cm), A is the area of the paddy plot (m^2), $CumLoss$ is the cumulative pesticide discharge during the period (% of applied mass) and A_{pp} is the pesticide application rate (mg m^{-2}).

The daily mass discharge of pesticides at the stations in the canal network was estimated by multiplying the daily flow volume by the daily concentrations in the stream. Concentrations below the detection limit were set to zero. The total pesticide losses from the monitoring stations covering different scales of paddy areas during the monitoring period were used to characterise the pesticide behaviour of the watershed.

Key factors affecting the pesticide fate and transport in the paddy watershed, such as hydrological data (rainfall) and management conditions, including the EWSC and water holding period (WHP), were investigated through an uncertainty analysis, the Monte Carlo simulation (MCS), a widely used method for probabilistic assessment and uncertainty analysis. The method involves random sampling from the distribution of inputs and successive model runs until a stable statistical distribution of outputs is obtained.[21] In this study, uncertainty analysis for pesticide discharge from a paddy plot was examined by MCS using the commercial available software package Crystal Ball version 7.1.[22]

The dymron and imazosulfuron discharges from paddy plot 1 were evaluated as a function of the rainfall and water management practices. Firstly, the surface drainage and herbicide discharge from plot 1 were calculated using the monitored water management scenario and the input data with a prescribed probability distribution by using MCS. Then the results were compared with monitored herbicide losses in order to evaluate the performance of model forecasting using MCS. The variability of the water management factors (EWSC and WHP) influencing pesticide losses from the paddy plot were then investigated using MCS with different water management scenarios.

The parameters for the dymron and imazosulfuron discharge from plot 1 using the MSC are presented in Table 3. The herbicides were applied on 1 May 1 and the simulation period was 46 days. Although the herbicide concentration in the paddy water seems to vary depending on water management, concentrations of dymron and imazosulfuron were assumed to be the same for all prescribed water scenarios and to be the same as with the monitoring data in plot 1 in this study because of the lack of monitoring data for the concentrations under different water scenarios. Probability distribution functions for the daily rainfall and total rainfall were specified on the basis of the data monitored over 3 years. The daily rainfall during the 46 day simulation period followed an exponential distribution with a rate exponent λ of 0.39. The maximum daily rainfall was 3.8 cm. The total rainfall during the period of 46 days was assumed to follow a uniform distribution, with observed maximum and minimum values of 11.8 and 10.1 cm respectively.

For the water management scenarios, the average daily $PERC$, $LSEEP$ and ET were obtained from the monitoring in plot 1. Since the probability distributions of these data were not known, they were assumed to follow a uniform distribution, with maximum and minimum values equal to the observed values plus 10% and minus 10% respectively. Note that the observed ET was set to 0 on rainy days, and the average values of 0.41 cm day^{-1} on non-rainy days. The maximum and minimum practised paddy water depths, $H_{\mathrm{w\,max}}$ and $H_{\mathrm{w\,min}}$, were set at 5.5 and 3.5 cm respectively, based on field observations of the entire watershed. This set-up resulted in an average water depth of 4.5 cm.

To create a datasheet for the daily water balance in the fields, firstly the amount of irrigation (IRR) was set to control the water depth level which fluctuated between $H_{\mathrm{w\,max}}$ and $H_{\mathrm{w\,min}}$. Thus, on days when the water level was below $H_{\mathrm{w\,min}}$, water was added to raise the water level to $H_{\mathrm{w\,max}}$. IRR was set to 0 on days with rainfall exceeding the total amount of $PERC$ and $LSEEP$. The daily EWSD was calculated from the water level corresponding to H_{gate} by Eqn (3). After that, daily rainfall data were added in the datasheet to calculate the amount of drainage (DR) using Eqn (2).

To analyse the probability of herbicide discharge from paddy plot 1, H_{gate} was set to 5.4 cm to create an EWSD of 0.9 cm, the same value as monitored in plot 1, and the WHP was set to 4 days as recommended by the pesticide labels (Table 3). To examine the effect of the EWSC, the WHP was assumed to be 4 days and the drainage gate was set to different heights ranging from 4 to 8 cm. This set-up created nine scenarios for the EWSC ranging from -0.5 (overflow drainage scenario) to 3.5 cm (Table 3). To examine the effect of the WHP, an EWSC of 0.5 cm was used, as the most frequent value obtained from field monitoring, and water scenarios with the WHP ranging from 0 to 15 days were defined (Table 3). The output parameters for the MCS were the average daily

Table 3. Parameters for pesticide discharge from the paddy field using Monte Carlo simulation

Parameter[a]	Symbol	Unit	Value	Distribution[b]	Comment
Pesticide					
Application rate					
Dymron	A_{pp}	$\mathrm{g\,m^{-2}}$	0.045	Point	Monitoring
Imazosulfuron	A_{pp}	$\mathrm{g\,m^{-2}}$	0.0085	Point	Monitoring
Paddy area (plot 1)	A	$\mathrm{m^2}$	3000	Point	Monitoring
Concentration in paddy water	C	$\mathrm{mg\,L^{-1}}$		Fig. 5	Monitoring
Rainfall					
Daily rainfall†	*RAIN*	$\mathrm{cm\,day^{-1}}$		Exponent (0.39) Maximum = 3.8 cm	Monitoring
Total rainfall for 46 days†	*RAIN*	cm		Uniform (10.1, 11.8)	Monitoring
Water management scenarios					
Daily percolation†	*PERC*	$\mathrm{cm\,day^{-1}}$	0.11	Uniform (Obs. ±10%)[c]	Monitoring
Daily lateral seepage†	*LSEEP*	$\mathrm{cm\,day^{-1}}$	0.22	Uniform (Obs. ±10%)[c]	Monitoring
Daily evapotranspiration†	*ET*	$\mathrm{cm\,day^{-1}}$	0.41	Uniform (Obs. ±10%)[c]	Monitoring
Maximum practised water depth	$H_{\mathrm{w\,max}}$	cm	5.5	Point	Monitoring
Minimum practised water depth	$H_{\mathrm{w\,min}}$	cm	3.5	Point	Monitoring
Average practised water depth	H_{w}	cm	4.5	Point	
1. *For probability analysis*					
Height of drainage gate	H_{gate}	cm	5.4	Point	
Excess water storage capacity	EWSC	cm	0.9	Point	Eqn (3)
Water holding period	WHP	days	4	Point	
2. *For investigation of EWSC*					
Height of drainage gate	H_{gate}	cm	4–8	Point	
Excess water storage capacity	EWSC	cm	−0.5–3.5	Point (9 scenarios)	Eqn (3)
Water holding period	WHP	days	4	Point	
3. *For investigation of WHP*					
Height of drainage gate	H_{gate}	cm	5	Point	
Excess water storage capacity	EWSC	cm	0.5	Point	Eqn (3)
Water holding period	WHP	days	0–15	Point (16 scenarios)	
Output data					
Average daily surface drainage	*DR*	$\mathrm{cm\,day^{-1}}$			Eqn (5)
Cumulative pesticide loss	*CumLoss*	% of applied mass			Eqns (2) to (5)

[a]† Parameter used for Monte Carlo simulation.

[b] To describe the distribution, lambda is used for exponential, the minimum and maximum values for uniform distribution.

[c] Maximum and minimum values: observed data plus and minus 10% respectively.

drainage and cumulative herbicide losses (*CumLoss*) calculated from the input parameters by Eqns (2) to (5). In this study, the MCS ran 10 000 computations.

3 RESULTS AND DISCUSSION

3.1 Watershed hydrology

3.1.1 *Water management in the paddy field*

The water management practices, including irrigation, drainage and water depth, applied on plot 1 in 2002 are shown in Fig. 2. The intermittent irrigation scheme (IntI) was clearly presented. Irrigation was applied according to the paddy water level. No significant paddy runoff occurred except during large rainfall events. The average daily precipitation, irrigation and drainage were estimated to be 0.25, 0.47 and 0.14 cm respectively. The average and maximum *ET* during the 46 day monitoring period in 2002 were about 4.1 and 8.4 mm day^{-1} respectively. For a typical Japanese paddy field it has been reported that the *ET* ranges up to 8 mm day^{-1}.[23]

The average value of the combined *PERC* and *LSEEP* was estimated to be about 3.3 mm day^{-1} from the water balance equation (Eqn 1) for plot 1. Owing to the flat paddy surface within a farm block, it was assumed that there was no water movement from one plot to an adjacent plot and that *LSEEP* mainly occurred through the short side field bund facing the secondary drainage canal. From the direct monitoring of the water balance in the drainage canal segments within farm blocks 1 and 2 during the monitoring periods for over 3 years, the average *LSEEP* from the

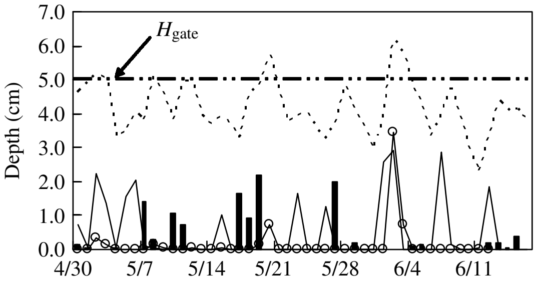

Figure 2. Monitored rainfall *RAIN* (▬), irrigation *IRR* (———), surface drainage *DR* (–o–) and paddy water depth H_{w} (- - - -) in plot 1 in 2002.

paddy fields into drainage canals was estimated to be 2.2 mm day^{-1}. Therefore, the remaining rate of 1.1 mm day^{-1} was assumed to be the *PERC* from the fields through the hardpan layer to the deep groundwater. It was reported that the rate of *LSEEP* is generally higher than the vertical percolation rate.[24,25] Such a low *PERC* rate seemed to be small compared with typical values reported for Japanese rice fields, which range from 5 to 30 mm day^{-1}.[23] However, the soil profile data (Table 4) showed a heavy clay layer of about 0.5 m depth at 1.0 m below the paddy field bed with a saturated hydraulic conductivity of 0.05–0.35 mm day^{-1}. The high clay content (49.5%) in the plough layer as compared with that of 14–37% for a typical paddy soil in Japan[26] is also one of the factors reducing the percolation rate in these rice fields.

The general water management practised in plot 1 (Fig. 2) has been introduced as the typical paddy plot water management in Japan.[27] About 5 cm of storage water in the plot with small daily changes in the first half and intermittent irrigation in the latter half of the growing season are practised. When the paddy plants grow around the maximum stooling stage to the ear premordia stage, a mid-summer drainage is conducted, that is, the ponded water in the paddy field is released and the field is allowed to remain dry for 8–10 days.[28] As a result of mid-summer drainage, the structure of the ploughed layer becomes denser and soil hardness is recovered sufficiently to allow machines to get into the fields for harvesting after ponded water release. The monitoring period of 46 days lasted from transplanting to mid-summer drainage, which covered the first half of the growing season.

The height of the drainage gate in plot 1 had been set to about 5 cm (Fig. 2), similarly to the typical field management in Japan as indicated by Watanabe.[27] The values of $H_{\mathrm{w\,max}}$ and $H_{\mathrm{w\,min}}$ were estimated at about 3.5 and 4.8 cm respectively on the non-rainy days. The EWSC or average EWSD estimated from non-rainy days was about 0.95 cm. The observed data showed that the paddy water level increased on rainfall events, and paddy runoff occurred following significant rainfall events exceeding about 1.5 cm day^{-1}. Two significant runoff events occurred: 0.8 cm day^{-1} on 20 May owing to a large rainfall and 3.4 cm day^{-1} on 3 June owing to excess irrigation.

The water storage depth has been known to be a key factor for many aspects of flood prevention in paddy fields.[29] However, its ability to control pollutant runoff from paddy fields from a water quality point of view has not been discussed. EWSD results from the height of the drainage gate, H_{gate}, and the paddy water level, and it therefore varies according to the farmers' practices. The measured (H_{gate}) and EWSD conducted in 296 paddy plots on a randomly selected non-rainy day (27 May 2005) followed normal distribution functions, with mean values of 5.2 and 0.5 cm for H_{gate} and EWSD respectively (Fig. 3). Standard deviations (STDV) of the normal distributions were 14.1 and 14.2 respectively for H_{gate} and EWSD. Note that the negative values of EWSD corresponded to overflow conditions such that the water level was higher than H_{gate}. The monitored data also showed that 113 out of a total of 296 surveyed plots had overflow drainage.

The type of water management scheme is another factor in pesticide runoff from paddy fields. It has been reported that a number of farmers in Japan are also office or factory workers.[1,5] These part-time farmers often apply a continuous irrigation and overflow drainage scheme (ContI) to save working time and sometimes because of insufficient irrigation pressure. As reported by Watanabe *et al.*,[9] application of a ContI scheme results in significant pesticide losses, especially during the early period after pesticide application. From direct measurement and interviewing farmers for 3 years it was established that about one-third of the paddy plots applied a ContI scheme where the measured average daily drainage was about 0.65 cm day^{-1}. The H_{gate} in plots with applied continuous irrigation and overflow drainage was often lower than about 4.5 cm. This fact caused a high pesticide concentration in the stream water, as discussed in the next section. In addition, the WHP 3–4 days after pesticide application was not well practised in plots applying the ContI scheme.

Table 4. Soil profile and physical properties

Depth (cm)	Organic carbon (%)	Particle size distribution (%) Sand	 Silt	 Clay	Hydraulic conductivity (mm day^{-1})
0–25	3.1	20.2	30.3	49.5	–
40–80	11.9	17.9	32.4	47.9	0.18–0.75
100–150	–	Heavy clay			0.05–0.35

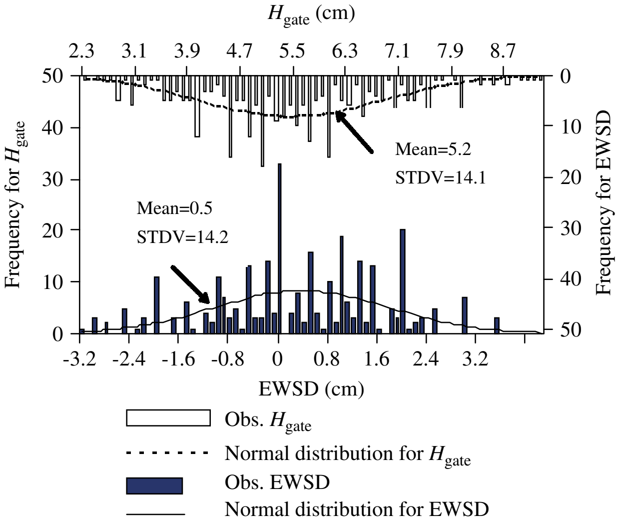

Figure 3. Probability distribution of H_{gate} and EWSD measured in 296 plots within the watershed on 27 May 2005.

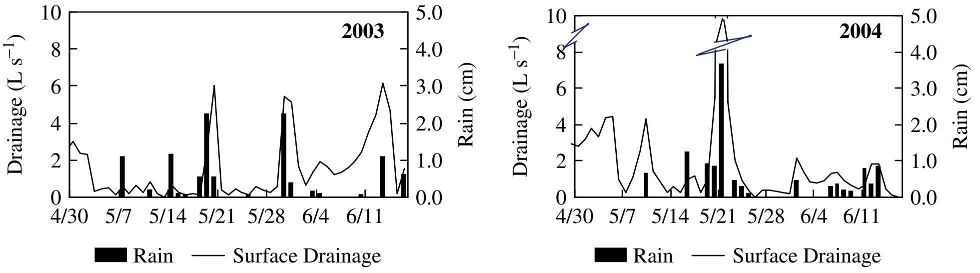

Figure 4. Surface drainage from farm block 2.

3.1.2 *Surface drainage from the farm block and canal discharge*

Figure 4 shows the surface drainage from paddy fields, including the runoff into the canal within farm block 2 in 2003 and 2004. Note that the surface drainage from the paddy fields was calculated from the water balance of the blocks. The average daily surface drainages estimated from the water balance in blocks 1 and 2 during the 46 day monitoring period were 0.27, 0.25 and 0.36 cm respectively for 2002, 2003 and 2004. The surface drainage from the paddy fields increased during significant rain events exceeding 1.5 cm day^{-1} (Fig. 4). This might be explained by the H_{gate} in some plots not being set high enough or the EWSCs not being large enough to store the rainfall water as discussed above. In some periods, the surface drainage from the farm blocks was not consistent with the rainfall events. This was probably due to the fact that some paddy plots in the farm blocks applied a ContI scheme. Increased surface drainage during the earlier period of 2003 and 2004 and the later period of 2003 did not correspond to the rainfall input (Fig. 4). For the earlier period, surface drainage increased owing to water release on transplanting so as to have an optimum ponding water depth for the seedlings. For the later period, the water release for the mid-summer drainage seems to be responsible for the increased surface drainage. From our observations over 3 years, the farmers started to release water from their fields for the mid-summer drainage period around 10 June and completed it around 20 June.

Discharge from the watershed monitored at the outlet (S10) responded to the observed rainfall pattern and the water management in the paddy fields (Fig. 5). Some peak flows occurred following significant rainfall events exceeding about 1.5 cm day^{-1}, and the lag time of the rainfall–discharge response was 12–24 h. In the earlier and later periods, the discharge increased without any significant rainfall input owing to the increase in paddy surface drainage during the transplanting and mid-summer drainage period, as already mentioned.

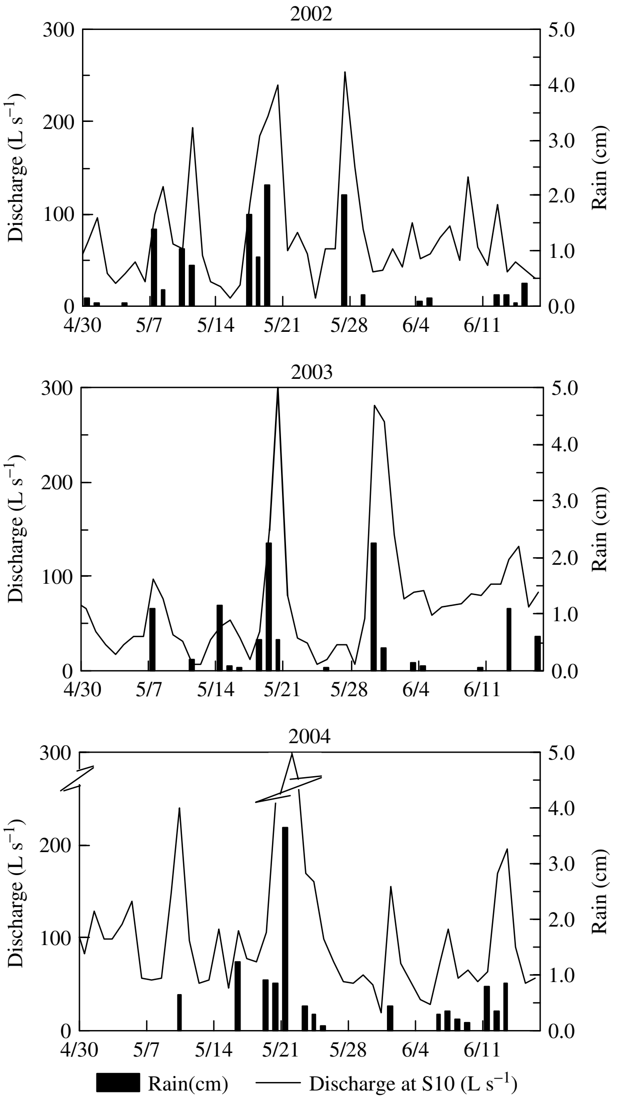

Figure 5. Discharge from the watershed monitored at S10.

3.1.3 *Hydrological balance within the watershed*

The hydrological balance in the watershed during the 46 day monitoring period in 2003 is shown in Table 5. The water balance in the paddy field compartment of 86 ha was evaluated on the basis of the water balance data in block 2 in 2003. The total or cumulative *IRR*, *DR*, *ET*, *PERC* and *LSEEP* during the 46 day period, expressed by depth of water, were estimated to be 35.5, 11.5, 18.9, 5.1 and 10.1 cm respectively. The average paddy water depth was assumed to be constant at 5 cm during the monitoring period. For the hydrological balance in the entire watershed, Q_{in} consisted of inflows monitored at S1, S2, S3 and S4 in the streams from Mt Tsukuba (Fig. 1) and Q_{out} was the discharge from the watershed into the Sakura River, measured at S10. The total or cumulative precipitation, the inflows from Mt Tsukuba, the irrigation supply from

Table 5. Hydrological balance in the watershed (2003)

Parameter	Symbol	Unit	Value[a]
Water balance in paddy field compartment of 86 ha			
Precipitation	*RAIN*	cm	10.1
Irrigation	*IRR*	cm	35.5
Drainage	*DR*	cm	11.5
Percolation	*PERC*	cm	5.1
Lateral seepage	*LSEEP*	cm	10.1
Evapotranspiration	*ET*	cm	18.9
Water balance in the whole watershed of 97 ha			
Precipitation	*RAIN*	cm	10.1
Inflow from Mt Tsukuba (at S1 to S4)	Q_{in}	cm	15.0
Irrigation supply from Lake Kasumigaura	Q_{lake}	cm	31.7
Percolation to groundwater	*PERC*	cm	5.1
Evapotranspiration	*ET*	cm	18.9
Outflow to River Sakura (at S10)	Q_{out}	cm	32.8
Circulation water (*IRR* – Q_{lake})	$Q_{cir.}$	cm	3.8

[a] Cumulative (or total) depth during 46 day monitoring period.

Lake Kasumigaura, *ET, PERC* and the outflow during the 46 day period, expressed in terms of depth of water, were 10.1, 15, 31.7, 18.9, 5.1 and 32.8 cm respectively (Table 5). From the hydrological balance calculations for the entire watershed area, 3.8 cm drainage water in the canals was estimated to be reused for irrigation through the pumping stations, corresponding to 32% of the paddy drainage water and 10.2% of the watershed discharge. Therefore, the cyclic irrigation system plays an important role in the rice paddy watershed from the point of view of improving both water use efficiency and water quality. Such an irrigation system has become popular in pipeline-irrigated areas in Japan.[30,31]

3.2 Herbicide concentrations

3.2.1 *Concentrations in the paddy water*

In 2002, the commercial herbicide Thoroughbred RX® SC, containing 17 g kg^{-1} imazosulfuron, 95 g kg^{-1} dymron, 66 g kg^{-1} clomeprop and 12 g kg^{-1} oxaziclomefone, was applied to plot 1 on 1 May, the same day of transplanting, at a rate of 5 kg ha^{-1}. Dymron and imazosulfuron were detected in the paddy water in high concentrations, while oxaziclomefone concentrations were negligible. Clomeprop was not determined. Dissipation of the two herbicides in the paddy water was quite similar. The maximum concentration of 653 and 113 µg L^{-1} respectively for dymron and imazosulfuron occurred on 2 May, on day 2 after pesticide application (APA), and then rapidly declined during the first 3 weeks (Fig. 6). Significant concentrations occurred during the first 2 weeks APA. Half-lives (DT_{50}) according to first-order kinetics were 6.2 and 6.7 days respectively for dymron and imazosulfuron. The corresponding values of DT_{90} (90% mass dissipation) were 21 and 22 days. The maximum oxaziclomefone concentration in the paddy water was 0.53 µg L^{-1}, which was very small compared with dymron and imazosulfuron. This was probably because oxaziclomefone has a very high sorption coefficient, K_{oc}, compared with dymron and imazosulfuron. The K_{oc} values for oxaziclomefone, dymron and imazosulfuron were 22 100, 3760 and 1 respectively.

WHP is one of the effective management practices for preventing herbicide runoff from paddy fields. However, the monitored results showed that the herbicides did not significantly decline during a period of 3–4 days (Fig. 6). This trend was also found for other rice herbicides such as mefenacet and pretilachlor,[32–35] thiobencarb,[36] carbofuran and molinate,[37] triclopyr and 2,4-D[38] and bensulfuron-methyl and azimsulfuron.[39] By field monitoring and a literature review, a previous study by the present authors suggested increasing the WHP up to 10 days according to the DT_{90} index instead of the current suggestion of 3–4 days in Japan.[9] The monitoring data in plot 1 also implied that increasing the WHP is important for reducing the herbicide discharge from the paddy fields. In the Sacramento Valley of California, the WHP has been applied for various periods depending on the active ingredient (up to 28 days for molinate and 30 days in the case of granular thiobencarb) and effectively inspected by the California Environmental Protection Agency.[40] However, in Japan, instructions for WHP seem to be available for farmers only on the pesticide label. Therefore, good water management practices including the appropriate WHP in the rice fields need to be given more attention and well practised through effective extension programmes.

3.2.2 *Concentrations in stream water*

A total of 16 herbicides were detected in the drainage water at stations, as shown in Figs 7 and 8. Dymron was detected at the highest concentration at all stations since it is a frequently used compound in this region and its application rate is relatively high compared with the other compounds. The peak concentration of dymron ranged from 33 to 108 µg L^{-1} in the secondary canal (at S6 and S7) and ranged from 25 to 50 µg L^{-1} in the main canal (at S10). Other herbicides detected in high concentrations were mefenacet, imazosulfuron and pretilachlor, with maximum concentrations ranging from 2 to 30 µg L^{-1}. The maximum concentration of mefenacet, one of the commonly used herbicides in Japan, in the secondary drainage canal was 2–3 times higher than the water quality advisory level of 9.0 µg L^{-1} for Japanese surface

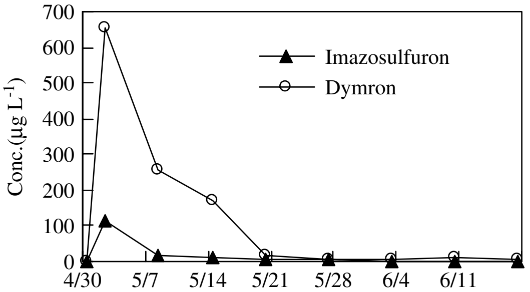

Figure 6. Herbicide concentrations in paddy water (plot 1 in 2002).

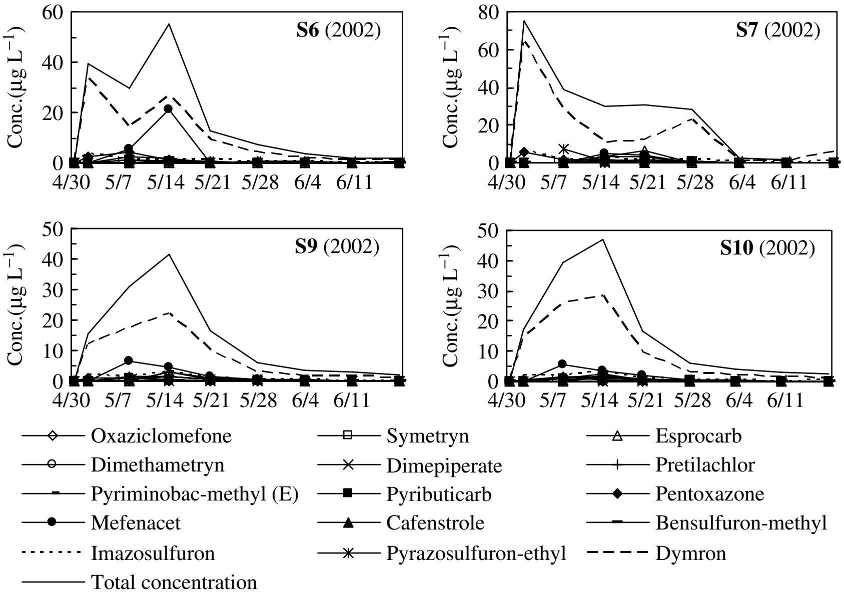

Figure 7. Herbicide concentrations in stream water at stations S6, S7, S9 and S10 in 2002.

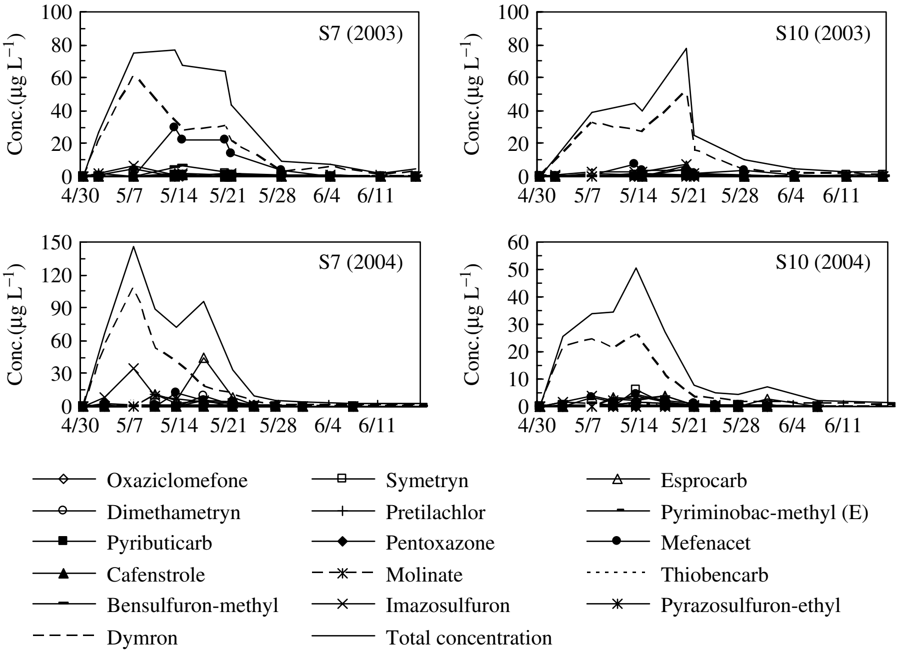

Figure 8. Herbicide concentrations in stream water at stations S7 and S10 in 2003 and 2004.

water.[41] On a farm block scale, peak concentrations of mefenacet were detected up to about 20 μg L^{-1} at S6 in 2002 and about 30 μg L^{-1} at S7 in 2003 (Figs 7 and 8). On a watershed scale, on account of dilution by the stream water, the maximum concentrations of mefenacet monitored at S10 ranged from 4 to 7.5 μg L^{-1} but still closely approached the water quality advisory level.

The maximum herbicide concentrations occurred mostly between 1 and 22 May, corresponding to the period of herbicide application and a short time afterwards (Figs 7 and 8). Thereafter the concentrations decreased rapidly in spite of the significant runoffs from the farm blocks. This was explained by the herbicide concentration in the paddy water declining drastically during the earlier period of about 2 weeks after application, as discussed in Section 3.2.1.

The times of the peak concentration varied among the monitoring stations. Within a station, the times and values of the peak concentrations varied between herbicides. These values and the times of the peak concentration of one herbicide also fluctuated year by year (Figs 7 and 8). These variations were probably due to variation in the timing of the application among the paddy plots. Communications with local farmers revealed that many growers chose the most suitable herbicide for their field and changed the product applied each year to prevent herbicide resistance problems. Most of the farmers start soil preparation in late April and transplant in early May. Depending on the selected products, the time of application varies from immediately after rice transplanting to about 3 weeks later. Surveying the farmers in farm block 2 of 19 plots in 2004 showed that the distribution of treatment dates followed a normal distribution, with the most frequent application date of 8 May and STDV of 2.8. Inao *et al.*[14] conducted a survey in 1996 and 1997 in a catchment of 2.71 km^2 having 338 paddy plots in the same prefecture (Ibaraki, Japan) and found the corresponding value on 13 May with an STDV of 3.5. In general, the herbicide application period in the region lasted for the first 3 weeks in May.

The total concentration in the stream water, which was the summed values of the concentrations of all detected herbicides, mostly peaked around 14 May (Figs 7 and 8) during the application season. The shape of the concentration curves was similar to those monitored in some other rivers in the region.[2,3,5,14] However, values of the peak concentrations in these rivers were only a few μg L^{-1}.

3.2.3 Pesticide losses

Figure 9 shows the daily and cumulative herbicide losses as a percentage of the mass applied in plot 1 in 2002. Cumulative losses were estimated to be 8.8 and 7.7% of the applied mass respectively for dymron and imazosulfuron, and mainly occurred following the two runoff events on 2 and 3 May (days 3 and 4 APA), as shown in Fig. 2. The herbicide runoff in plot 1 (Fig. 9) may not be representative for all plots in the watershed, but it implies that field practice and water management by farmers during the recommended WHP is important for controlling herbicide losses because of the high concentrations in this period.

The total herbicide losses from the watershed monitored at S10 were estimated to be about 5.3, 6.9 and 7.0 kg respectively for 2002, 2003 and 2004. Significant daily herbicide losses occurred, corresponding to the peak flow after significant rainfall events (Fig. 10). More than 70% of the total loss of the detected herbicides was discharged from the watershed from 7 to 22 May, corresponding to the application period. In the later period after 3 weeks from application, the herbicide runoff was small due to the low herbicide concentrations in the paddy water, despite the significant water discharge as discussed in the previous section. The same trend in herbicide loading was seen in stations S8 and S9 in the main canal.

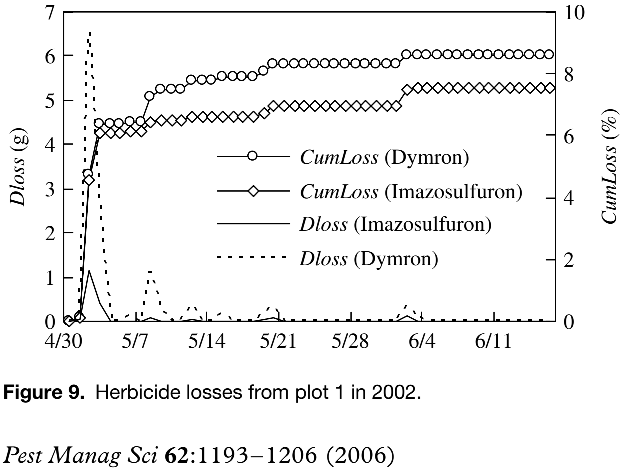

Figure 9. Herbicide losses from plot 1 in 2002.

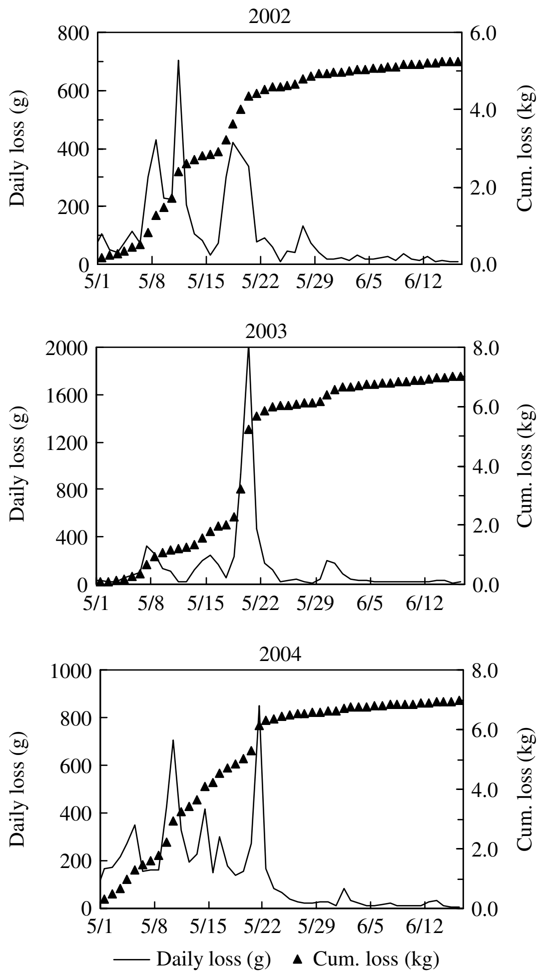

Figure 10. Herbicide losses from the watershed monitored at S10.

Owing to the lack of data for herbicide use in the entire watershed, the data surveyed from block 2 in 2004 were assumed to be applicable to the entire watershed for the estimated percentage of herbicide discharge. In spite of the uncertainty in the pesticide input, a general picture of pesticide loading from the watershed was presented and compared with published results. The total amount of active ingredients (AI) included in the commercial herbicides used in block 2 varied from about a few percent to roughly 20%, which is about 0.3–2 kg AI ha^{-1} according to the recommended application rates. Supposing that these herbicides were applied to the 86 ha of paddy field in the studied watershed, then, a preliminary estimate of the total active ingredient that applied to the watershed was from about 26 kg to 172 kg in the case of the application rates ranging from about 0.3 to 2 kg AI ha^{-1} as above. The average cumulative loss from the watershed monitored at S10 over 3 years was about 6.4 kg (Fig. 10), which corresponds to about 25% to 3.7% of the preliminary estimated applied mass of 26 kg to 172 kg. The total herbicide losses from block 2 monitored at S7 in

2004 was about 0.72 kg, or 12.8% of the total applied ingredient of 5.6 kg. Sudo *et al.*[4,13] reported that annual losses of the herbicides from the watersheds studied ranged from 0 to 13.0%, depending on the pesticide. Another study showed that pesticide losses ranged from 8 to 23%.[5]

Cyclic irrigation is a factor affecting pesticide behaviour in the watershed. As discussed in the previous section, it was estimated that, during the 46 day period, about 3.8 cm of drainage water in the canal was reused for irrigation, corresponding to a 10.2% discharge from the watershed (Table 5). Assuming that the herbicide concentrations in this reused water were equal to the concentrations monitored at the outlet of the watershed (S10), an average of 0.71 kg of AI was estimated to be reused in the paddy field.

The total loss of the herbicides detected relative to the paddy area is shown in Fig. 11. A regression analysis based on the data for 2002 showed a good correlation between the total losses and the paddy area ($R^2 = 0.994$). The estimated herbicide loss per unit area based on the monitored data in 2002 was about 0.059 kg ha^{-1}. The values calculated from the monitoring data at the outlet of the watershed (S10) in 2003 and 2004 were respectively 0.082 and 0.081 kg ha^{-1} greater than in 2002. This was probably because discharges from the watershed in 2003 and 2004 were larger than in 2002, and partly due to the total concentrations of the detected herbicides monitored at the outlet of the watershed (S10) in 2002 being lower than in 2003 and 2004 (Figs 7 and 8). The cumulative discharges from the watershed at S10 over the monitoring period were 327×10^3, 372×10^3 and 390×10^3 m^3 respectively for 2002, 2003 and 2004. However, the total rainfall during the 46 day monitoring period showed small fluctuations of 11.6, 10.1 and 11.8 cm respectively for 2002, 2003 and 2004. These data indicated that the discharge from the watershed and herbicide concentrations in the stream water fluctuated year by year, depending on the irrigation management and probably on the pesticide use in the watershed.

3.3 Uncertainty analysis

The distribution of the cumulative herbicide losses (*CumLoss*) and average daily paddy runoff from plot 1 using MCS is shown in Fig. 12. The distribution is normal for surface drainage and positive skewed normal for *CumLoss*. The mean standard errors were 0.075 and 0.05 for the *CumLoss* of dymron and imazosulfuron respectively. The mean value of the average daily surface drainage was 0.16 cm day^{-1} (Fig. 12), compared with the monitored value of 0.14 cm day^{-1}. The 50 percentile cumulative herbicide losses obtained by MCS were 12.6 and 6.5% respectively for dymron and imazosulfuron. The *CumLoss* of dymron and imazosulfuron monitored in plot 1 were 8.8 and 7.7% respectively (Fig. 9). Therefore, the MCS can provide a reasonable forecast of the herbicide discharge from a paddy plot using the probability of the input data.

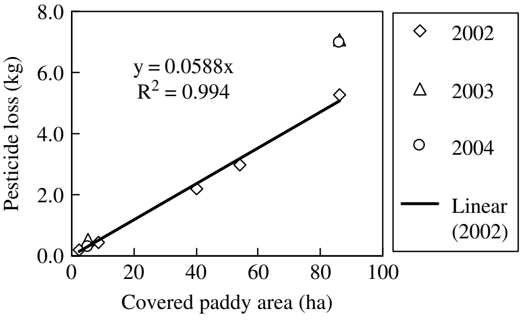

Figure 11. Characteristic of herbicide losses responding to covered paddy area.

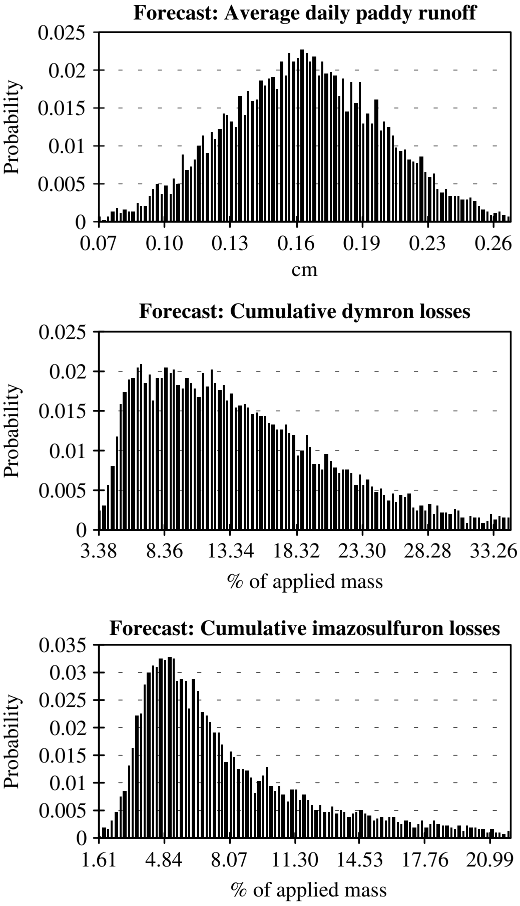

Figure 12. Distribution of average daily paddy runoff and cumulative herbicide loss from plot 1 using Monte Carlo simulation (EWSC = 0.9 cm).

The mean values of the cumulative herbicide loss (*CumLoss*) and daily paddy runoff from plot 1 calculated by MCS versus EWSC are shown in Fig. 13. The drainage and both herbicide losses indicated similar patterns. Note that the effect of changes in the herbicide concentrations, depending on the management scenarios, was not considered. However, the slope of the curves clearly showed the effect of EWSC on herbicide losses. In the case of negative EWSC (overflow drainage scenario) resulting from the H_{gate} being set up below 4.5 cm, herbicide discharges were more than 30 and 50% respectively

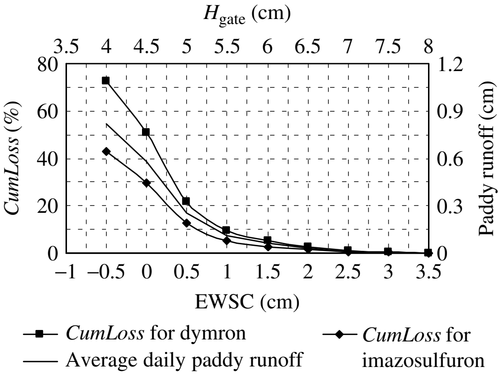

Figure 13. Relationship between excess water storage capacity (EWSC) and herbicide losses.

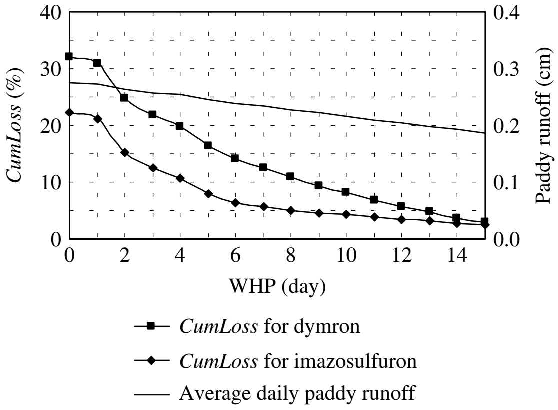

Figure 14. Relationship between water holding period (WHP) and herbicide losses.

for imazosulfuron and dymron. The average daily drainage for these cases was greater than 0.6 cm, similar to the 0.65 cm monitored from the plots using the ConI scheme, as discussed in the previous section. Reductions in *CumLoss* were quite rapid when EWSC increased from 0 to 1 cm, but slowed down thereafter. With an EWSC of 1 cm, *CumLoss* was still considerable at about 5.5 and 10% for imazosulfuron and dymron respectively, and the average daily drainage was about 0.14 cm, quite similar to that monitored in plot 1. An EWSC greater than 2 cm, resulting in less than 3% herbicide loss, should be recommended as the appropriate water management technique. With a typical practised paddy water depth of about 4–5 cm,[42] setting the EWSC greater than 2 cm requires the drainage gate to be set up higher than 6.5 cm.

Figure 14 presents the effect of WHP on the reduction in *CumLoss* and daily surface drainage. The results were calculated with MCS using the monitored rainfall data and typical water management in Japan,[42] with H_{gate} set to 5.5 cm and the average practised paddy water depth assumed to be 4.5 cm, resulting in an EWSC of 0.5 cm (Table 3). Reduction in the daily paddy runoff was a steady and slow decline, with an average rate of 0.006 cm per day of WHP. However, the reduction in *CumLoss* with change in WHP from 0 to about 1 week was fast compared with that with the longer WHP owing to the high concentration in the paddy water during the first week. With the recommended WHP of 3–4 days, the *CumLoss* was still more than 10 and 20% for imazosulfuron and dymron respectively. However, a WHP of 10 days would reduce the pesticide losses to less than 5%.

4 CONCLUSIONS

Discharge from a canal network and pesticide runoff responded to significant rainfall events. The key factors of pesticide runoff from this paddy field watershed were considered to be the significant pesticide concentrations shortly after the pesticide application and runoff from the paddy fields following significant rainfall events. For controlling herbicide runoff from paddy fields in a monsoon region such as in Japan, the recommended best management practices or good agricultural practices are summarised as follows:

- For the paddy plot level it could be an application of an intermittent irrigation scheme with a high drainage gate and shallow paddy water depth practice creating an EWSC greater than 2 cm to store rainfall water in the paddy field during significant rainfall events. A water holding period for at least 10 days according to the DT_{90} index is recommended instead of the current regulation of 3–4 days.
- For the watershed level the cyclic irrigation system may play an important role in the reduction in pesticide discharge from the watershed into the river. The combination of good water management practices in the rice fields and small-scale water cycling is probably one of the alternative methods for best management practice in a paddy watershed to reduce the pesticide runoff into the river.

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