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Simulation of mefenacet concentrations in paddy fields by an improved PCPF-1 model

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Abstract: An improved simulation model (PCPF-1) has been evaluated for the prediction of the fate of mefenacet in an experimental paddy field. This model simulates the fate and transport of pesticide in paddy water and the top 1 cm of paddy soil. Observed concentrations of mefenacet in the paddy water and the surface soil exponentially decreased from their maximum concentrations of 0.70 mg litre^{-1} and 11.3 mg kg^{-1}, respectively. Predicted mefenacet concentrations both in the water and surface soil were in excellent agreement with those measured during the first 2 weeks after herbicide application, but concentrations in paddy water were appreciably overestimated thereafter. The model simulated mefenacet losses through runoff, percolation and degradation to be respectively 41.9%, 6.4% and 57.3% of applied, and the mass balance error was about −6%. The model simulation implied that drainage and seepage control, especially shortly after application when herbicide concentrations are high, is essential for preventing pesticide losses from paddy fields. In focusing on pesticide concentrations in this early period the PCPF-1 model can be a beneficial tool for risk assessment of pesticide losses and in the evaluation of agricultural management for reducing pesticide pollution associated with paddy rice production.

Keywords: paddy field; herbicide; runoff; simulation model; mefenacet

1 INTRODUCTION

About 40% of the agricultural land in Japan is cultivated for paddy rice (1.94 million ha) and this receives about 50% of the total amount of agricultural pesticides.[1] However, a typical paddy field in Japan is susceptible to herbicide runoff since the chemical is applied directly to paddy water. Pesticide losses from paddy fields range from a few percent to more than 50% of that applied depending on the water management.[2,3] Monitoring river systems in Japan has detected several herbicides commonly used in paddy fields[3–6] and these concentrations appear to have adverse effects on the aquatic ecosystem.[6] Establishing best-management practices for controlling pesticide discharge from paddy is very important in protecting aquatic ecosystems.

Mefenacet [2-(1,3-benzothiazol-2-yloxy)-*N*-methylacetanilide] is used pre- and early post-emergence, mainly in transplanted rice to control grass weeds.[7] Mefenacet was one of the top three selling pesticides in Japan from 1993 to 1997,[8] and has often been detected in surface waters during the rice crop season.[3–6] Mefenacet concentrations in water range from more than 1000 µg litre^{-1} in the paddy plot[9,10] to about 30 µg litre^{-1} in drainage canals and tributary streams in an extensive paddy fields watershed,[3,4,6,11] and about 17 µg litre^{-1} in a main river (Kiose river).[5] These concentrations peaked during the month of May following the application period. The guideline value for surface-water quality for mefenacet in Japan is 9 µg litre^{-1}.[12] Mefenacet has been reported to be phytotoxic to several algal species, with 72 h EC$_{50}$ values of 250–670 µg litre^{-1}.[6]

The use of simulation models to determine the predicted environmental concentration (PEC) has become the basis for assessing the potential environmental risk within the regulatory and registration process. In the European Union (EU), advisory groups such as FOCUS and MED-RICE have been working to facilitate the standard tiered approach for pesticide risk assessment in rice production using mathematical models.[13] Currently, a number of simulation models for pesticides used in paddy rice production are available. The RICEWQ model has been used under US and EU conditions for simulating PEC in surface water as well as in ground water.[14,15] PCPF-1[16,17] and PADDY[18] have been validated for paddy rice conditions in Japan.

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Simulation models can be also a powerful tool for analysing the mechanisms of pesticide fate and transport phenomena, and evaluating best management practices in controlling pesticide discharges from paddy fields. In paddy fields, the timing and amount of rainfall after pesticide application greatly influence pesticide runoff.[11] The importance of water management in this has been elucidated through monitoring and simulation of different management scenarios using both PADDY and PCPF-1.[17,18] The PCPF-1 model has been validated using measured parameters and input variables obtained from a monitoring study investigating the fate of pretilachlor, an acetamide herbicide commonly used in rice paddy.[16,17] Since the initial version, PCPF-1 has been modified to improve data accessibility, the analytical scheme for the output data and model predictability.

The objectives of this paper were to evaluate the improved model for the prediction of mefenacet concentrations in an experimental paddy field, and to assess the fate of mefenacet so as to minimize water pollution associated with rice production.

2 MATERIALS AND METHODS

2.1 Simulation model for pesticide concentrations in paddy fields (PCPF-1)

PCPF-1 simulates the pesticide fate and transport in two compartments: the paddy water and surface soil. The paddy water compartment is assumed to be a completely mixed reactor having variable water depths. The paddy surface soil compartment is also assumed to be a completely mixed reactor, with a constant depth of 1.0 cm. According to Takagi *et al.*,[19] this surface layer of paddy soil is aerobic even when submerged, so that pesticide degradation occurs under oxidative conditions.

Considering the paddy field environment, the conceptual pesticide fate scenario used for the model is shown in Fig. 1. Since the publication of the previous PCPF-1 model,[16] several points have been modified to improve model performance. For the water balance component, input data for lateral seepage and a new module for the estimation of evapotranspiration are included. For the pesticide fate calculations, photochemical degradation is simulated using solar radiation instead of UV-B radiation, as measured values of the former are more readily available.

The PCPF-1 model consists of three governing equations including water balance in the paddy water compartment and mass balances of pesticide both in paddy water and in the surface soil layer. The daily water balance within the water compartment is given by

$$\frac{\mathrm{d}h_{\mathrm{PW}}}{\mathrm{d}t} = \mathrm{RAIN} + \mathrm{IRR} - \mathrm{DRAIN} - \mathrm{LSEEP} - \mathrm{PERC} - \mathrm{ET_C} \qquad (1)$$

where h_{PW} is the depth of water in paddy field (cm), t is time (day), RAIN is the average rainfall rate during dt (cm day^{-1}), IRR is the rate of irrigation water supply (cm day^{-1}), DRAIN is the surface drainage or overflow rate (cm day^{-1}), LSEEP is the rate of lateral seepage out of the plot through levees or bunds (cm day^{-1}), PERC is the rate of vertical percolation (cm day^{-1}) and $\mathrm{ET_C}$ is the rate of evapotranspiration (cm day^{-1}).

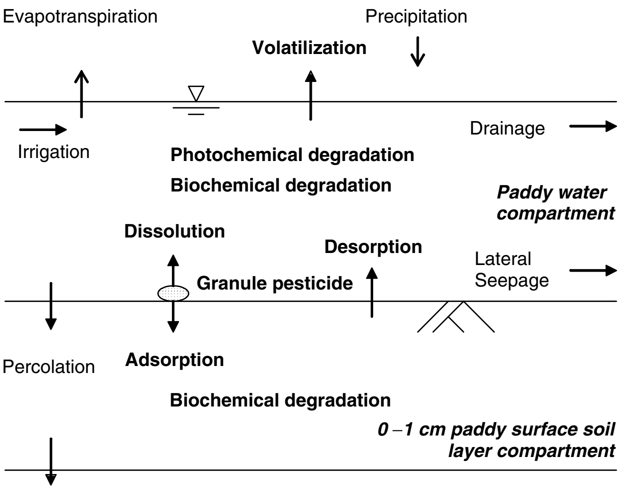

Figure 1. Conceptual pesticide fate in a paddy-rice field.

The equation governing the mass balance of pesticide in the paddy water compartment is expressed by

$$\begin{aligned}\frac{\mathrm{d}C_{\mathrm{PW}}}{\mathrm{d}t} &= k_{\mathrm{DISS}}\,(C_{\mathrm{SLB}} - C_{\mathrm{PW}}) + \frac{1}{h_{\mathrm{PW}}}\left[C_{\mathrm{PW}}\frac{\mathrm{d}h_{\mathrm{PW}}}{\mathrm{d}t}\right]_{\mathrm{DISS}} \\ &\quad + \frac{1}{h_{\mathrm{PW}}} d_{\mathrm{PSL}}\rho_{\mathrm{b-PSL}} k_{\mathrm{DES}} C_{\mathrm{S-PSL}} \\ &\quad + \frac{1}{h_{\mathrm{PW}}}\mathrm{IRR}\; C_{\mathrm{W-IRR}} - \frac{1}{h_{\mathrm{PW}}}(\mathrm{DRAIN} \\ &\quad - \mathrm{LSEEP} - \mathrm{PERC})C_{\mathrm{PW}} \qquad (2) \\ &\quad - \frac{1}{h_{\mathrm{PW}}}\mathrm{k}_{\mathrm{L-A}} C_{\mathrm{PW}} \\ &\quad + (-k_{\mathrm{PHOTO}} f_{\mathrm{US}} R_{\mathrm{S-a}}(1 - f_{\mathrm{R-ab}} t) \\ &\quad - k_{\mathrm{BIOCHEM-PW}})C_{\mathrm{PW}} - \frac{1}{h_{\mathrm{PW}}}\frac{\mathrm{d}h_{\mathrm{PW}}}{\mathrm{d}t}C_{\mathrm{PW}}\end{aligned}$$

where C_{PW} is pesticide concentration in paddy water (mg litre^{-1}), k_{DISS} is the first-order rate constant of pesticide dissolution in water (day^{-1}), C_{SLB} is the solubility of pesticide in water (mg litre^{-1}), d_{PSL} is the depth of the paddy surface soil layer (cm), $\rho_{\mathrm{b-PSL}}$ is the bulk density of the paddy surface soil layer (g cm^{-3}), k_{DES} is the first-order rate constant for pesticide desorption from the paddy surface soil layer (day^{-1}), $C_{\mathrm{S-PSL}}$ is the pesticide concentration in the paddy surface soil (mg kg^{-1} dry soil basis), $C_{\mathrm{W-IRR}}$ is the pesticide concentration in irrigation water (mg litre^{-1}), $k_{\mathrm{L-A}}$ is the pesticide mass transfer coefficient from paddy water to atmosphere (cm day^{-1}) and $k_{\mathrm{BIOCHEM-PW}}$ is the first-order rate constant of biochemical degradation in paddy water (day^{-1}).

A modified algorithm for photochemical degradation accounts for the attenuation by plant growth of the sunlight entering the paddy water (factor $f_{\mathrm{R-ab}}$).

$R_{\mathrm{S-a}}$ is the daily solar radiation ($\mathrm{kJ\,m^{-2}}$) above the rice canopy and k_{PHOTO} is the first-order rate coefficient of photochemical degradation with respect to the cumulative UV-B radiation ($\mathrm{m^2\,kJ^{-1}}$) as measured in ambient or laboratory conditions.

Similarly, the pesticide mass balance in the paddy surface soil layer is considered as the following equation:

$$\begin{aligned}
\frac{\mathrm{d}C_{\mathrm{S-PSL}}}{\mathrm{d}t} &= k_{\mathrm{d-PSL}}\, k_{\mathrm{DISS}}\,(C_{\mathrm{SLB}} - C_{\mathrm{PW}}) \\
&\quad + k_{\mathrm{d-PSL}} \left[\frac{C_{\mathrm{PW}}}{d_{\mathrm{PSL}}}\frac{\mathrm{d}(d_{\mathrm{PSL}})}{\mathrm{d}t}\right]_{\mathrm{DISS}} \\
&\quad + \frac{k_{\mathrm{d-PSL}}}{(\theta_{\mathrm{Sat-PSL}} + \rho_{\mathrm{b-PSL}} k_{\mathrm{d-PSL}})}\frac{1}{d_{\mathrm{PSL}}}\mathrm{PERC} \\
&\quad \times \left(C_{\mathrm{PW}} - \frac{1}{k_{\mathrm{d-PSL}}} C_{\mathrm{S-PSL}}\right) \\
&\quad - \frac{k_{\mathrm{d-PSL}}}{(\theta_{\mathrm{Sat-PSL}} + \rho_{\mathrm{b-PSL}}\, k_{\mathrm{d-PSL}})} \\
&\quad \times \rho_{\mathrm{b-PSL}}\, k_{\mathrm{BIOCHEM-PSL}}\, C_{\mathrm{S-PSL}} \\
&\quad - \frac{k_{\mathrm{d-PSL}}}{(\theta_{\mathrm{Sat-PSL}} + \rho_{\mathrm{b-PSL}} k_{\mathrm{d-PSL}})} \\
&\quad \times \rho_{\mathrm{b-PSL}}\, k_{\mathrm{DES}}\, C_{\mathrm{S-PSL}} - \frac{C_{\mathrm{S-PSL}}}{d_{\mathrm{PSL}}}\frac{\mathrm{d}(d_{\mathrm{PSL}})}{\mathrm{d}t}
\end{aligned} \tag{3}$$

where $k_{\mathrm{d-PSL}}$ is the soil adsorption coefficient of the pesticide ($\mathrm{litre\,kg^{-1}}$), $\theta_{\mathrm{sat-PSL}}$ is the volumetric saturated water content ($\mathrm{cm^3\,cm^{-3}}$), $k_{\mathrm{BIOCHEM-PSL}}$ is the first-order rate constant of the biochemical degradation of the pesticide ($\mathrm{day^{-1}}$) and the subscript PSL refers to the paddy surface soil layer.

The model program was coded using Visual Basic® for Applications in Microsoft Excel®. The input data consist of 23 measured parameters, together with the daily water balance of the paddy water and local meteorological data. For the new version, the calculation time step was changed from one day to one hour in order to improve predictability. A module for pesticide mass balance was also included in the new version in order to facilitate further data analysis. This module calculates and prepares a datasheet for the pesticide in paddy water and 1 cm surface soil, pesticide losses by surface runoff, percolation and seepage, loss by degradation including photochemical and biochemical degradation as well as volatilization. Data are presented as percentage of applied mass on a daily basis. In the mass balance, all of the herbicide is assumed to dissolve in paddy water. Herbicide losses though percolation below the paddy surface soil layer were calculated by multiplying the rate of percolation by the herbicide concentration in soil water as given by the partitioning coefficient and the concentration in the paddy surface soil layer. In addition, the cumulative mass balance error (see Table 4) was calculated as the difference between the applied mass and the sum of the dissipated masses and the mass of the residue in paddy water and surface soil layer.

The Macro program calculates and automatically creates output data and figures in a Microsoft Excel® file. Detailed explanations of the model development and evaluation are given in Watanabe and Takagi.[16,17]

2.2 Modifications of PCPF-1 model

2.2.1 Modification of the photochemical degradation algorithm

In the previous version,[16,17] the photochemical degradation process in paddy water was assumed to follow first-order kinetics (Eqn 4):

$$\frac{\mathrm{d}C_{\mathrm{PW}}}{\mathrm{d}t} = (-k_{\mathrm{PHOTO}}\, R_{\mathrm{UVB-b}})C_{\mathrm{PW}} \tag{4}$$

where k_{PHOTO} is the first-order rate coefficient of photochemical degradation with respect to the cumulative UV-B radiation ($\mathrm{m^2\,kJ^{-1}}$) and $R_{\mathrm{UVB-b}}$ is the daily UV-B radiation ($\mathrm{kJ\,m^{-2}}$) below the rice canopy, which was measured about 10 cm above the paddy water. The value of the parameter k_{PHOTO} is determined by laboratory measurements of pesticide dissipation under UV-B radiation.

In order to avoid the need to measure the UV-B radiation to use as an input variable, the model was improved by calculating the photochemical degradation of pesticide in paddy water using the solar radiation above the rice canopy, which is usually available from the weather station database. At first, the relationship between daily UV-B radiation above and below rice canopy was defined as

$$\frac{R_{\mathrm{UVB-a}} - R_{\mathrm{UVB-b}}}{R_{\mathrm{UVB-a}}} = f_{\mathrm{R-ab}}\, t \tag{5}$$

where $R_{\mathrm{UVB-a}}$ and $R_{\mathrm{UVB-b}}$ are the daily UV-B radiation above and below the rice canopy, respectively ($\mathrm{kJ\,m^{-2}}$), $f_{\mathrm{R-ab}}$ is the slope of the fitted line obtained from the relative difference of the radiation below and above the rice canopy which accounts for the light attenuation by the growing rice crop, and t is time (day). The UV-B radiation below the rice canopy is calculated from the solar radiation above rice canopy as

$$R_{\mathrm{UVB-b}} = f_{\mathrm{US}} R_{\mathrm{S-a}}(1 - f_{\mathrm{R-ab}}\, t) \tag{6}$$

where $R_{\mathrm{UVB-b}}$ is the UV-B radiation below the rice canopy, f_{US} is the fraction of the UV-B radiation over solar radiation below the rice canopy, and $R_{\mathrm{S-a}}$ is the solar radiation above the rice canopy. A new expression for the photochemical degradation process in paddy water is derived from the above equations as

$$\frac{\mathrm{d}C_{\mathrm{PW}}}{\mathrm{d}t} = -k_{\mathrm{PHOTO}}\, f_{\mathrm{US}} R_{\mathrm{S-a}}(1 - f_{\mathrm{R-ab}}\, t)C_{\mathrm{PW}} \tag{7}$$

Photochemical degradation is now calculated from Eqn (7) using solar radiation data. Note that the parameter k_{PHOTO} is still determined experimentally with respect to UV-B radiation; however, parameter modification with respect to solar radiation can be derived using Eqn 4.

2.2.2 Simulation model for crop evapotranspiration in a paddy field

The previous version used measured values of crop evapotranspiration (ET_C) as input. However, the current version has a program and data sheet for the estimation of ET_C in a paddy field when the required meteorological data are available. We applied the FAO Penman–Monteith method, since FAO recommends this as a consistent and globally valid standard to calculate the water requirement of crops. Full details of this method with the crop coefficient approach and the procedures for determining the parameters, algorithms, recommended values and units are available.[20] The Penman–Monteith equation used for 24 h calculations of reference evapotranspiration, ET_O, using mean daily data can be simplified as follows:

$$ET_o = \frac{0.408\Delta(R_n - G) + \gamma \frac{900}{T + 273} U_2(e_s - e_a)}{\Delta + \gamma(1 + 0.34U_2)} \tag{8}$$

where ET_O is the reference crop evapotranspiration (mm day^{-1}), R_n is the net radiation at the crop surface (MJ m^{-2} day^{-1}), G is the soil heat flux (MJ m^{-2} day^{-1}), T is the average air temperature (°C), U_2 is the wind speed measured at 2 m height (m s^{-1}), e_s is the saturation vapour pressure (kPa), e_a is the actual vapour pressure (kPa), $(e_s - e_a)$ is the saturation vapour pressure deficit (kPa), Δ is the slope of the vapour pressure curve (kPa °C^{-1}), γ is the psychrometric constant (kPa °C^{-1}), and 900 is the conversion factor for 24 h calculations.

For the standard conditions of field management having the optimum water scheme, and without considering constraints such as water stress, diseases and pests, the ET_C from a rice paddy can be estimated by the crop coefficient approach, whereby the effects of the various weather conditions are incorporated into ET_O in Eqn (8) above and the influence of the crop characteristics into the K_C coefficient:

$$ET_C = K_C ET_O \tag{9}$$

The model program was also coded by Visual Basic for Applications in Microsoft Excel® within the PCPF-1 structure. A detailed description of the development of ET_C simulation program for application to the pollutant model is presented in Vu *et al.*[21]

2.3 Model simulation

The PCPF-1 model was executed with observed data from the experimental paddy field at the National Institute of Agro-Environmental Sciences (NIAES) in Tsukuba, Japan from May 13 to July 15 1998.[17] Measured daily data of precipitation, irrigation, drainage, lateral seepage, percolation and paddy water levels (cm) during 64 days were input via Eqn (1) into a data worksheet of the PCPF-1 model. Also, daily maximum, minimum and average air temperatures (°C), daily average of wind speed at 2 m above ground (m s^{-1}), relative humidity (%) and solar radiation (MJ m^{-2}) for the ET_C simulation were input in a worksheet. At first, the ET_C simulation was executed with the input parameters set according to Table 1, and then PCPF-1 was executed. Parameter values for pesticide fate processes (Table 2) were determined through laboratory experiments conducted at NIAES by the same procedures as previously.[17,19] The model offers the options of two-phase first-order kinetic rate coefficients for pesticide desorption and biochemical degradation in order to account for the slower degradation at lower concentrations.[16,17] Mefenacet behaviour was simulated using a single-phase first-order rate coefficient for both the dissipation and biodegradation processes.

Table 1. Input parameters for estimating evapotranspiration

Variety	Nihonbare
Planting date (yy/mm/dd)	98/05/07
Total cropping period (days)	125
1st stage period (days)	21
2nd stage period (days)	35
3rd stage period (days)	42
4th stage period (days)	27
Crop coefficient for initial stage, K_{Cini}	1.1
Crop coefficient for mid-season stage, K_{Cmid}	1.2
Crop coefficient for late season stage, K_{Cend}	0.9
Elevation of field above sea level (m)	20
Latitude of field (decimal degrees)	36.167
Location: Northern or Southern Hemisphere	N

Table 2. Input parameters for PCPF-1 model simulation

	Unit	Value
Paddy water compartment		
Solubility of the pesticide	mg litre^{-1}	4.0
1st-order dissolution rate constant	day^{-1}	0.239
1st-order desorption rate constant (Phase 1)	day^{-1}	0.0626
Mass transfer coefficient of pesticide volatilization	m day^{-1}	3.50E-06
1st-order photochemical degradation rate constant	m^2 kJ^{-1}	0.0062
1st-order biochemical degradation rate constant	day^{-1}	0.0941
Pesticide concentration in irrigation water	mg litre^{-1}	0
1st-order desorption rate constant (Phase 2)	day^{-1}	0.0626
Phase intercept concentration for desorption	mg litre^{-1}	1
Factor of light attenuation by crop	day^{-1}	0.0162
Paddy soil compartment		
1st-order biochemical degradation rate constant (Phase 1)	day^{-1}	0.0343
Equilibrium soil adsorption coefficient	litre kg^{-1}	24.07
1st-order biochemical degradation rate constant (Phase 2)	day^{-1}	0.0343
Phase intercept concentration for biochemical degradation	mg litre^{-1}	0.1

2.4 Field monitoring of mefenacet concentrations in a paddy field

Environmental conditions required for the model and pesticide concentrations in paddy water and paddy surface soil were monitored at an experimental rice paddy plot at NIAES in 1998. Following paddy field preparation and water ponding,[22] the soil was puddled and levelled by several passes of a rotary tiller under a few centimetres of water-ponding conditions. After a few days, 17-day-old rice seedlings (*Oryza sativa* L. cv. Nihonbare) were transplanted at 16 × 30 cm spacing on May 8 1998. The granular herbicide Zark-D®, containing 35 $g\,kg^{-1}$ mefenacet, 15 $g\,kg^{-1}$ daimuron and 1.7 $g\,kg^{-1}$ bensulfuron-methyl, was applied at 40.8 $kg\,ha^{-1}$ (1.43 kg mefenacet ha^{-1}) on a 9.1 × 9.1 m paddy plot on May 13.

Environmental variables monitored were paddy water depth, irrigation, drainage, evapotranspiration, percolation, precipitation, and solar and UV-B radiation above and below the rice canopy. Evapotranspiration during the experiment was monitored by daily measurement of the water level in an adjacent lysimeter with an equivalent plant density of four rice plants. In addition, meteorological data for minimum, maximum and average daily temperature, wind speed and relative humidity were obtained from the Weather Data Acquisition System of NIAES for the estimation of the ET_C. The paddy surface soil (0–1 cm) was a light clay soil having a particle size distribution of 46.7% sand, 19.4% silt and 33.9% clay, with soil $pH(H_2O)$ of 5.2 and organic carbon content of 1.83%. Paddy water and 1 cm surface paddy soil were sampled at 1, 3, 7, 14, 21, 28, 35 (water only), 42 and 49 days after herbicide application at about 3 pm before the daily irrigation.

2.5 Measurement of mefenacet concentrations

2.5.1 Extraction

Paddy soil samples (20 g fresh weight, two replicates) in an Erlenmeyer flask with acetone (100 ml) were sonicated (3 min), shaken (30 min) and filtered (Kiriyama paper no. 5A, 60 mm diameter). The soil was extracted again with acetone (50 ml) and the filtrates combined. The filtrate was concentrated in a rotary evaporator (40–45 °C) until all the acetone was removed. The remaining aqueous solution (about 10 ml) was mixed with sodium chloride solution (50 g $litre^{-1}$; 30 ml) in a separating funnel, to which a flask hexane rinse (30 ml) was added. After partitioning, the hexane layer was passed through a silicone-treated filter (Whatman 1PS); the aqueous layer was extracted similarly a second time. The combined hexane extracts were then evaporated to dryness using an automated evaporator (Zymark TurboVap II; 50 °C; 25 min), and the residue dissolved in acetone (10 ml). Analysis by gas chromatography gave a recovery of 96.9% with a coefficient of variation of 3.0% for three replications. The limit of detection was 10 $\mu g\,kg^{-1}$ dry soil.

Water samples were filtered through a glass-fibre filter paper (Whatman GF/B) and pH was adjusted to 6.5–7.0. Sep-Pak C18 cartridges were preconditioned by washing with acetone (5 ml), methanol (5 ml) and distilled water (8 ml). The water samples were passed through the conditioned cartridges using a pump (Waters) at a flow rate of 10 ml min^{-1}. The volumes passed through the cartridges for samples taken during the 1st and 2nd weeks were 200 ml, during the 3rd and 4th weeks 600 ml, and during the 5th to 7th weeks 1000 ml. The cartridges were then dried over a vacuum manifold (Waters) for about 30 min and then eluted with acetone (5 ml). Before GC analysis, samples were dried in the automated evaporator and redissolved in acetone (from 5 ml to 1 ml depending on the sample concentrations). The recovery for mefenacet was 91.7%, with a coefficient of variation of 3.82% for three replications. The limit of detection was 0.05 $\mu g\,litre^{-1}$.

2.5.2 Gas chromatography

The Hewlett Packard HP 5890 Series II gas chromatograph had a nitrogen–phosphorus detector and splitless injection onto a capillary column (HP-50+ with 50% Ph Me silicone gum) with an inside diameter of 0.53 mm, length 15 m and film thickness 1 µm. The temperatures were 250 °C, 280 °C and 280 °C for oven, injector port and detector, respectively. The GC was equipped with an automatic sampler with injection volume 2 µl.

3 RESULTS AND DISCUSSION

3.1 Evaluation of photochemical degradation algorithm

The monitored solar and UV-B radiation above and below the rice over the 64 days from May 13 to July 8 in 1998 and 1999 in the experimental paddy plots (Table 3) were used to determine the parameters f_{R-ab} and f_{US}. The cumulative values of solar radiation and UV-B radiation above and below the rice canopy indicated that consistency in variation between two years of data was low, probably because of the effects of the accuracy of the measurement device and its placement on paddy water below the rice canopy (Table 3). Heisler *et al.*[23] reported that relative irradiance (I_r = beneath-trees/above-canopy irradiance) for UV-B may range from 0.01–0.02 beneath dense forest canopies to about 0.4 in the shade of a single tree. The I_r values applied to paddy water for 1998 varied from 1.0 in the earlier period to 0.08 by 64 days after transplanting; however, that in 1999 varied from 1.0 to only 0.35. Relative irradiance above and below the rice canopy strongly depends on the shading conditions associated with the position of the measurement device and the crop growth.

For the simulation, the default value of the fraction of the UV-B radiation over solar radiation, f_{US}, was determined to be 1.232×10^{-3} from the average value of the 1998 and 1999 observations (Table 3). The

Table 3. Monitoring results of solar and UV-B radiation above and below rice canopy in 1998 and 1999

Parameter	1998	1999
Cumulative solar radiation above rice canopy (MJ m^{-2})	817	1029
Cumulative solar radiation below rice canopy (MJ m^{-2})	663	784
Cumulative UV-B radiation above rice canopy, (kJ m^{-2})	1256	1290
Cumulative UV-B radiation below rice canopy (kJ m^{-2})	604	1057
f_{R-ab} for solar radiation (R^2)[a]	0.0087 (0.629)	0.0085 (0.850)
f_{R-ab} for UV-B radiation (R^2)[a]	0.0159 (0.958)	0.0079 (0.711)
f_{US} above rice canopy (R^2)[b]	1.496×10^{-3} (0.836)	1.192×10^{-3} (0.783)
f_{US} below rice canopy (R^2)[b]	0.929×10^{-3} (0.928)	1.309×10^{-3} (0.928)

[a] f_{R-ab} is the slope of fitted line of Eqns (5)–(7) and its R^2.

[b] f_{US} is the fraction of the daily UV-B over daily solar radiation in Eqns (6) and (7).

average f_{US} value was compromised to a default value because we were not able to determine the cause of the variability of the f_{US} value either by the measurement position (above or below the rice canopy) or by the year, as data were limited. Nonetheless, the f_{US} values obtained from the data of May 13 to July 8 provided by the National Institute of Environment (NIES) located about 2 km from the experimental plots in NIAES were 1.290×10^{-3} in 1998 and 1.032×10^{-3} in 1999, comparable to our monitoring data. Similarly, the corresponding default value of the factor accounting for the light attenuation by the rice crop, f_{R-ab}, was 0.0103 averaged over 1998 and 1999 (Table 3).

The sensitivity of the model to the parameters affects its performance; however, the photochemical degradation of mefenacet was only minor. The relative mean errors (RME) of daily herbicide concentrations in paddy water calculated on changing the k_{PHOTO} value by +10% and −10% from the default value ($0.0062\,m^2\,kJ^{-1}$) were −1.7% and +1.8%, respectively. The corresponding value for the surface soil was less than 1%. For pretilachlor tested with the data of Watanabe and Takagi,[17] corresponding values were less than 1% for both compartments.

The model sensitivities to the variable parameters, f_{US} and f_{R-ab}, were tested by changing the input value by +50% and −50% from the default value. For f_{US}, the RME in daily herbicide concentrations in paddy water using 150% of default value was −8.0% and that for 50% of default value was +9.9%. Corresponding values for pretilachlor were −1.4% and +1.5%, respectively. The prediction error seemed to depend upon the sensitivity of the pesticide to photochemical degradation. The photochemical degradation rate constant of mefenacet is more than seven times larger than that of pretilachlor.[17] The relative mean absolute error (RMAE) in daily herbicide concentrations in paddy water for new algorithms to that for the previous version was 0.2% for pretilachlor and 2.4% for mefenacet. Corresponding RMAE values for paddy soil were less than 1% for both herbicides. The new program is now able to simulate using solar radiation data, which is easily available at weather stations, yet maintaining comparable accuracy to the previous program.

3.2 Simulation of evapotranspiration in a paddy field

The simulated crop evapotranspiration (ET_C) in a paddy field was evaluated using measurements over the pesticide monitoring period. However, the FAO Penman–Monteith estimation was found to be subject to both random and systematic errors.[21] The maximum difference between the observed and estimated cumulative ET_C values is up to 25% for Japanese rice cultivation and this method may require careful calibration according to the regional conditions and varieties.[21] Therefore, in this simulation, the input parameters for the crop coefficients of the rice growing stage (K_{Cini}, K_{Cmid} and K_{Cend}; Table 1) were adjusted according to the monitoring data following the FAO guidelines.[20] In order to avoid appreciable systematic errors, the values of K_C were adjusted to correspond to the minimum values of the bias between prediction and observation.[21] From the lysimeter measurements, the estimated value of the crop coefficient for the initial stage, K_{Cini}, was 1.1, the same value as indicated by the FAO guidelines. The corresponding value for the mid-season stage, K_{Cmid}, was 1.2, about 3% different from that indicated by the FAO guidelines. For the random errors of the daily ET_C estimation, it is suggested that the FAO Penman–Monteith method is more reliable when calculating cumulative ET_C over periods of more than 7 days.[21] During 57 days of this monitoring period (May 13 to July 8), cumulative ET_C estimated by the FAO method was only 0.5% greater than the observed value. The RMAE in daily herbicide concentrations in paddy water simulated by PCPF-1 using estimated and observed ET_C was 4% and that of paddy surface soil was 0.2%. Using uncalibrated K_C values recommended by the FAO guidelines, corresponding RMAE in daily herbicide concentrations were up to 5.6% for other scenarios.[21] Detailed descriptions for the evaluation of the program to estimate evapotranspiration in different lysimeter experiments have been presented.[21]

3.3 Simulation of mefenacet concentrations in a paddy field

The PCPF-1 model simulation was evaluated with the field monitoring of mefenacet concentrations in the paddy water and 0–1 cm surface paddy soil layer in

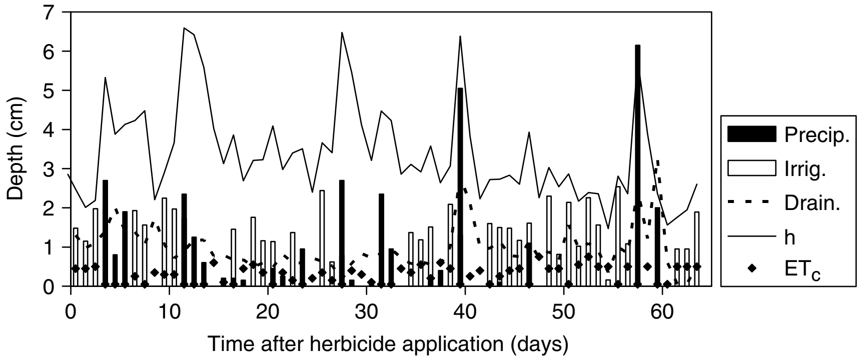

Figure 2. Observed daily precipitation (Precip.), irrigation (Irrig.), drainage (Drain.), paddy water depth (*h*) and evapotranspiration (ET_C) during the monitoring period.

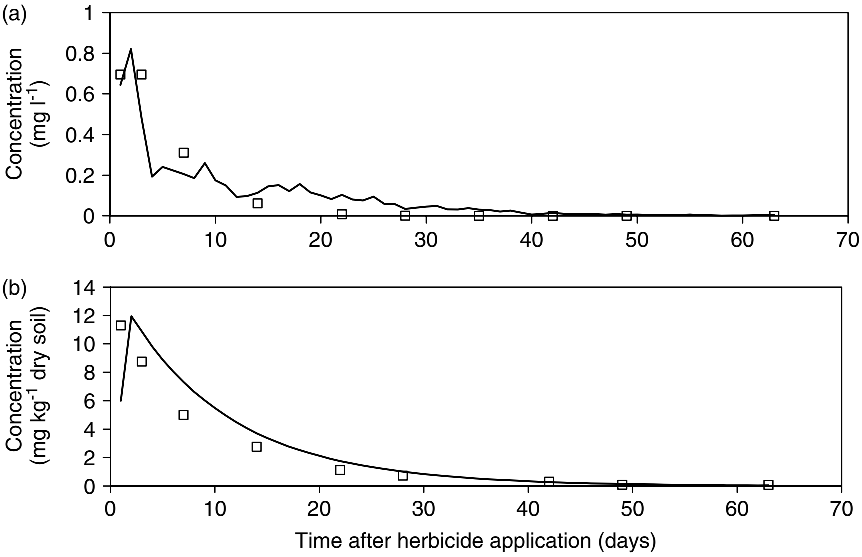

Figure 3. Simulated (———) and observed (□) mefenacet concentrations in (a) paddy water and (b) 0–1 cm surface soil.

the experimental paddy field from May 13 through July 15 in 1998.[17] In this monitoring, appreciable amounts of vertical edge flow through the concrete border and some leak of paddy water through the plastic border interface to the drainage basin were observed. Since these water paths were assumed to have negligible interaction with the soil matrix, these water losses were combined with surface drainage for the purposes of pesticide monitoring. The average amounts of precipitation, irrigation, surface drainage, percolation and measured evapotranspiration during the monitoring period were 0.52, 0.93, 0.96, 0.20, and 0.29 cm day^{-1}, respectively (Fig. 2). The paddy water depth fluctuated between about 2 and 6 cm due to irrigation and rainfall, and averaged 3.4 cm.

Water management depends on hydrological condition, soil physical characteristics and farmers' management practices. For typical Japanese paddy, evapotranspiration ranges up to about 0.8 cm day^{-1} and percolation rate ranges from 0.5 to 3.0 cm day^{-1}.[24] Surface drainage is highly dependent on farm practices and Watanabe and Maruyama[25] observed about 0.35 cm day^{-1} as an average value. Mizutani,[26] reporting on typical water balance in Japanese paddy, gave average depths of precipitation, irrigation, surface drainage, deep percolation, lateral seepage and evapotranspiration during the irrigation period of 120 days of 0.75, 1.50, 0.55, 0.30, 0.90 and 0.50 cm day^{-1}, respectively.

Observed concentrations of mefenacet in paddy water and surface soil layer decreased exponentially from their respective maximum concentrations of 0.70 mg $litre^{-1}$ and 11.3 mg kg^{-1} (Fig. 3). The half-life (DT_{50}) and 90% dissipation time (DT_{90}) values obtained using first-order kinetics for the paddy water were 4.3 and 14.4 days, respectively, and corresponding values for the paddy surface soil layer were 8.2 and 27.1 days, respectively.

Similar results were obtained from previous 3-year monitoring studies.[10] Ishii *et al.*[9] also reported that maximum mefenacet concentrations ranged from 0.93 mg $litre^{-1}$ for Zark-D granule formation to 0.24 mg $litre^{-1}$ for Act granule formation. Mefenacet DT_{50} and DT_{90} values for their experiment were 3.4 and 9.7 days, respectively, for paddy water, and 12.0 and 41.0 days, respectively, for paddy soil. Observed pretilachlor dissipation in 1998 showed comparable behaviour.[17]

For the simulation of mefenacet concentrations by the improved PCPF-1 model, the drastic decline of mefenacet concentrations in paddy water during the first week as the result of significant rain events (2.7 cm on day 4 and 1.9 cm on day 6 after herbicide application) was well simulated. Increases in water level (Fig. 2) correspond to the decline of simulated mefenacet concentrations (Fig. 3), which implies that the model successfully simulated the dilution effects of those appreciable rainfall events. The relative mean absolute error (RMAE) of the simulated pesticide concentrations in paddy water to those measured during the first 2 weeks was 0.24. However after the 3rd week, the model significantly overestimated the amounts observed. On the exponential scale, the predicted concentration decreased linearly to 0.0035 mg litre^{-1} at the end of the simulation, whereas observed concentrations declined more rapidly and remained at the level of 10^{-4} mg litre^{-1} after the 4th week. The model prediction for the mefenacet concentration in the paddy surface soil layer was good. The RMAE for the observed values were 0.39 for the first 2 weeks. The model prediction after the 3rd week was also excellent. Observed and predicted mefenacet concentrations in paddy surface soil layer showed an exponential decrease to 0.07 mg litre^{-1} at the end of the simulation.

In soil, mefenacet is strongly adsorbed and shows little movement.[7] In sterile aqueous buffer solutions, mefenacet undergoes slow hydrolysis at all pH levels; however, in natural water it is degraded more rapidly.[7] According to Fajardo *et al.*,[10] half-lives of mefenacet in paddy water ranged from 3.3 to 4.1 days and those in 0–1 cm surface paddy soil ranged from 9 to 11 days during 3 years of observation. Mefenacet dissipation in paddy field conditions has been successfully simulated using the PADDY model with parameters obtained in the laboratory and from the literature.[10]

The comparisons between observed and predicted mefenacet concentrations for paddy water and surface soil layer calculated by the previous (daily simulation) and modified versions (hourly simulation) were analysed statistically (Table 4). Bias is the measure of the systematic error in the model prediction, whereas variance measures random error and mean absolute error evaluates the model performance affected by both systematic and random error.[27] The RMAE of the simulated pesticide concentrations to those observed in paddy water and surface soil were 0.34 and 0.39, respectively. This comparison indicated that model performance with respect to systematic error was greatly improved. A possible reason for this improvement was that hourly simulation may give better estimation for samples taken at certain times of the day (3 pm for this simulation), whereas daily simulation corresponds to the daily average of the pesticide concentrations, which was different from our sampling procedure. The mass balance error was improved to be −5.96%, as compared to the previous version of 10.18%. Although random error in the prediction of the mefenacet concentrations in soil slightly increased for the modified version, in general its performance was improved.

3.4 Mefenacet fate and transport in a paddy field

The fate and transport of mefenacet in a paddy field were evaluated with the dataset prepared by the pesticide mass balance module. The potential risks that lead to significant pesticide losses from paddy fields and the management factors controlling such losses were assessed. After 63 days in 1998 (Fig. 4), mefenacet residues in paddy water and 1 cm surface paddy soil were calculated to be 0.04% and 0.27%, respectively, and the cumulative losses by surface drainage or runoff and percolation were 41.9% and 6.4%, respectively. The loss by degradation,

Table 4. Statistical comparison of previous (daily simulation) and modified (hourly simulation) PCPF-1 model performance

Model compartment	Previous		Modified	
	PW	PSL	PW	PSL
Bias	−0.038	−0.375	−0.013	0.113
Relative bias	−0.217	−0.112	−0.076	0.034
Variance	0.019	3.845	0.007	4.374
Relative variance	0.599	0.343	0.007	4.387
Mean absolute error	0.081	1.122	0.227	0.390
Relative MAE	0.460	0.335	0.340	0.390
R^2	0.794	0.759	0.946	0.721
Mass balance error (%)	10.18		−5.96	

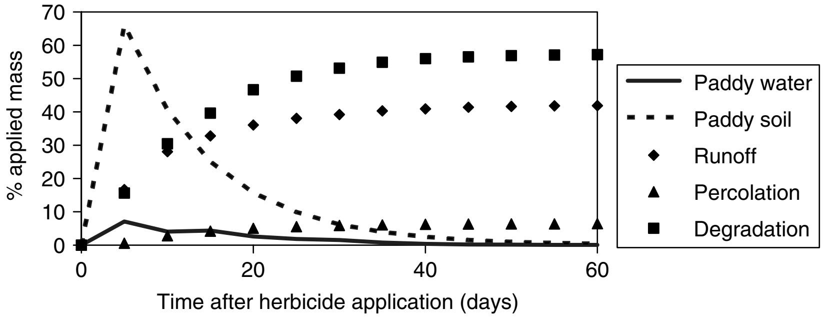

Figure 4. Mefenacet distribution during the monitoring period for pesticide residues in paddy water and in paddy surface soil layer, and pesticide losses by surface drainage, percolation and degradation.

including photochemical and biochemical degradation and volatilization, accounted for 57.3%. Note that the mass balance error still remains at about −6% as shown in Table 4 and that the seepage loss was not calculated in this simulation.

Mefenacet loss via surface drainage was the major loss process in this experimental plot. However, note that the drainage includes the amount of vertical edge flow through the concrete border and some leak of paddy water through the plastic border interface to the drainage basin. For the simulation of pretilachlor dissipation by PCPF-1, 52% was predicted to be lost through surface drainage and 6.4% through percolation.[17] In the case of continuous irrigation overflow drainage, the runoff loss of mefenacet from a 0.3 ha experimental paddy plot was 38% of applied mass,[28] and simetryn and molinate losses simulated by the PADDY model were 41% and 62% of applied mass, respectively.[18] However, the measured runoff loss of mefenacet from 268 ha of paddy fields accounted for 14.5% of applied.[4] For pesticide fate and transport, the timing of transport factors such as runoff and seepage when pesticide concentrations are high is also important. In this experiment, more than 80% of mefenacet runoff occurred within first 2 weeks and this phenomenon contributed to the drastic decline in the herbicide concentration during the initial period. Herbicide concentrations in paddy fields during the first 2 weeks are appreciable, and even small amounts of drainage or seepage during this period would result in considerable herbicide losses. Other studies also revealed that, for water management by continuous irrigation overflow, more than 80% of mefenacet runoff[28] and more than 90% of total molinate and simetryn runoff[18] occurred within the first week after application. In California, rice growers are required to hold water on their paddy fields following application of rice pesticide; water-holding periods for molinate, granular thiobencarb (Bolero®) and liquid thiobencarb (Abolish®) are 28, 30 and 19 days, respectively.[29] Although recent seepage water management still requires further effort to meet their water quality performance goal, implementation of the water-holding requirement successfully improved local stream water quality.[30]

As with surface drainage, controlling percolation rate is also important for preventing excess leaching of agricultural chemicals. Miao *et al.*[14] simulated that approximately 19% of applied cinosulfuron leached through the vadose zone in a typical Italian rice production having a percolation rate of 0.23 cm day^{-1}. The percolation rate in the paddy field depends on the soil physical characteristics as well as field management practices such as puddling. In Japan, puddling is a common land preparation which makes the plough layer soft and level to facilitate transplanting, and this reduces percolation.[22] Therefore, puddling also seems to be important for the control of non-point source pollution.

For the scenario with 0.23 cm day^{-1} percolation, the RICEWQ-VADOFT model simulated that 80.5% of applied cinosulfuron was degraded, the runoff loss being less than 1%. Needless to say, the chemical characteristics of the applied pesticide also affect its fate. PCPF-1 simulated that 57.3% of applied mefenacet, having relatively high K_d and low water solubility, was lost by degradation. In contrast, pretilachlor, having a lower K_d value and higher water solubility, was estimated to dissipate chemically by only 26% with a runoff loss of more than 60%.[17] Nevertheless, the amount of pesticide dissipation by various processes of degradation could be increased when runoff and leaching losses were minimized, as in a field operated by a closed system approach. Therefore considering good agricultural practice, drainage and seepage control, especially in earlier period with high pesticide concentrations, seems to be essential for preventing pesticide losses from paddy field and improving surface and ground water quality.

4 CONCLUSIONS

Predicted mefenacet concentrations both in paddy water and surface soil were in excellent agreement with those observed during the first 2 weeks after herbicide application, although significant overestimation occurred for concentrations in paddy water thereafter. The model simulated mefenacet losses through runoff, percolation and degradation to be respectively 41.9, 6.4 and 57.3% of applied, and the mass balance error was about −6%. The improvements in the photochemical degradation algorithm, the estimation of evapotranspiration and the pesticide mass balance module enhanced data accessibility and analytical capability. The performance of the model was also improved by applying a 1 h simulation interval.

For good agricultural practice, control of both drainage and seepage, especially in the early period with high pesticide concentrations, seems to be important for preventing pesticide losses from paddy fields. In focusing on pesticide concentrations in this early period following application, the PCPF-1 model can be a beneficial tool for the risk assessment of pesticide losses and the evaluation of agricultural management for reducing pesticide pollution associated with paddy rice production.

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