JARQ 42 (1), 13 – 21 (2008) http://www.jircas.affrc.go.jp

REVIEW
Simulation Models of Pesticide Fate and Transport in Paddy Environment for Ecological Risk Assessment and Management

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Abstract

Rice production is one of the major non-point sources of pesticide pollution in Japan. Monitoring of pesticide concentrations in river systems detected a number of herbicides commonly used in paddy fields, and these concentrations may appear to have adverse effects on the aquatic ecosystem. In this paper, two mathematical models developed in Japan, the PADDY and PCPF models for simulating the fate and transport of pesticides in a paddy environment are introduced. These models have been validated with observed data from laboratory experiments and field monitoring studies. In addition, the application of a mathematical model (RICEWQ) modified in Europe for high tier risk assessment in paddies is provided. We applied the PADDY and PCPF models to controlling pesticide runoff losses from paddy fields and to ecological risk assessment in the aquatic environment. The recommendation from model simulations for reducing pesticide runoff from paddy fields are 1) application of an intermittent irrigation scheme with a high drainage gate and 2) application of a longer water holding period after pesticide application. In order to establish a realistic assessment and management procedure for environmentally-friendly rice production, it is important to develop and validate mathematical models adapted to paddies in the Asian region.

Discipline: Agricultural chemicals
Additional key words: exposure assessment, mathematical model, pesticide runoff, water management

Introduction

Located in the east end of the temperate Asian monsoon region, Japan had a total agricultural land area of 4.7 million hectares in 2006, and paddy fields under rice cultivation accounted for 1.7 million ha. Most of the paddy fields are treated with herbicides, fungicides and insecticides which are applied during the crop season accordingly. Therefore, rice production is one of the major non-point sources of pesticide pollution. The typical paddy field in Japan is susceptible to herbicide runoff since the chemical is applied directly onto paddy water. Pesticide runoff losses from paddy fields range from a few percent to more than 50% of the applied amount depending on the water management[15,27]. Monitoring of pesticide concentrations in river systems in Japan detected a number of herbicides commonly used in paddy fields[9,20,22,27], and these herbicides may appear to have adverse effects on the aquatic ecosystem[22].

In this paper, two mathematical models developed in Japan, the PADDY and PCPF models for simulating the fate and transport of pesticide in a paddy environment are introduced. In addition, the application of a mathematical model (RICEWQ) modified in Europe for high tier risk assessment in paddy fields is provided. Then, we applied

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Received 4 January 2007; accepted 6 August 2007.

the PADDY and PCPF models to controlling pesticide runoff losses from paddy fields and to ecological risk assessment in the aquatic environment.

Simulation of pesticide fate and transport in a paddy environment using the PADDY model series

1. PADDY model

The PADDY model calculates pesticide concentration in paddy water and surface soil by considering pesticide behavior and average water balance in a paddy field as shown in Fig. 1[6,7]. The pesticide behavior is controlled by the interaction of the following processes: the dissolution of pesticide from granules into paddy water, the adsorption and desorption of pesticide between paddy water and soil, the runoff, the leaching and the volatilization of pesticides from paddy water, and the degradation of pesticide in paddy water and soil. The mass balance equations in paddy water and surface soil are expressed in terms of the kinetics of fate and transport processes. In the model, steady state was applied in the water balance. The model program was coded using Microsoft Visual Basic® based on Windows. The model input data consist of environmental conditions in the paddy field (water balance and soil properties) and pesticide parameters (physicochemical properties, equilibrium constants and rate constants).

To validate this model, a field experiment was carried out in a paddy field in Tokyo from June 24 through July 22 in 1991 using two herbicides (molinate and simetryn)[6]. The concentrations of these herbicides were measured in paddy water and surface soil. The predicted results by the model were in a good agreement with the experimental ones (Fig. 2). The PADDY model can also estimate the contribution of the fate and transport processes in herbicide dissipation[6].

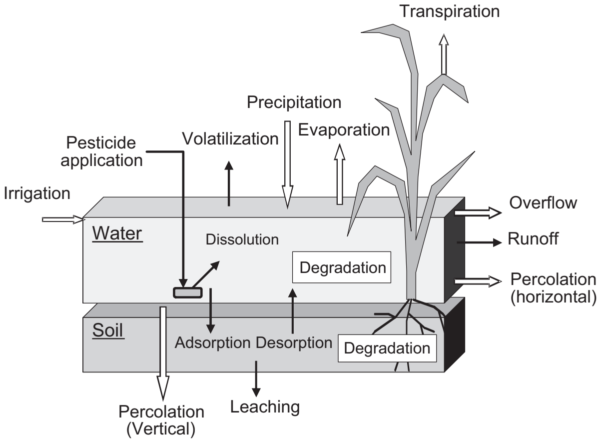

Fig. 1. Conceptual water movement and pesticide fate and transport in paddy field of the PADDY model[6,7]
→: Pesticide transport. ⇨: Water movement.

2. PADDY-2 model

An improved version of the PADDY model (PADDY-2) was evaluated to estimate the behavior of pesticides more accurately by considering daily water balance in a paddy field with site-specific environmental conditions[7]. For the water movement processes in paddy fields, the PADDY-2 model considers irrigation, precipitation, evaporation, transpiration, drainage (overflow), and both vertical and horizontal percolation as shown in Fig. 1. In the PADDY-2 model, daily water depth and outflow rate can be calculated by the water balance equation.

To validate the PADDY-2 model, field studies were performed in the experimental paddy fields (40 m^2) at National Institute for Agro-Environmental Sciences under two different water management conditions in 1996 using molinate and simetryn. In the first treatment the water depth was maintained at about 4 cm by occasionally supplying irrigation water without outflow. The other treatment involved continuous irrigation where the outlet of

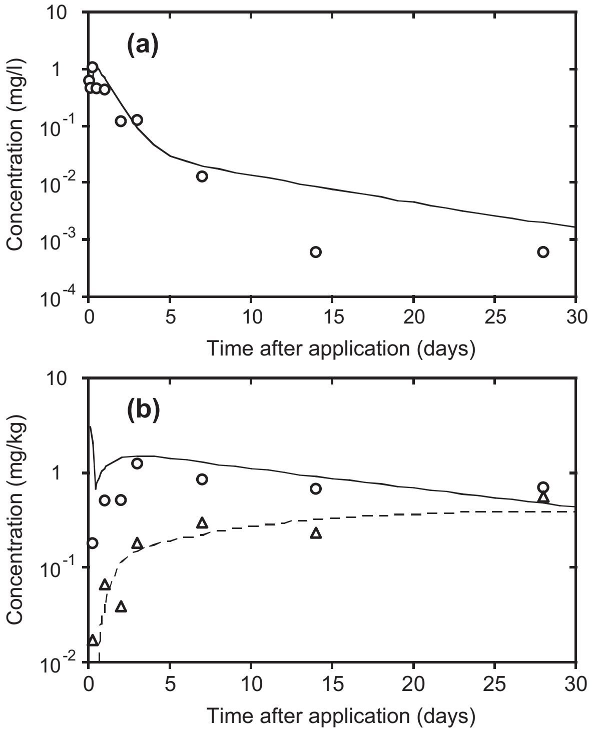

Fig. 2. Comparison between simulated and measured concentration of simetryn in paddy field[6]
(a): Concentration in paddy water. —: Simulated, ○: Measured. (b): Concentration in soil. 0–2 cm: — ; Simulated, ○ ; Measured. 2–4 cm: ⋯ ; Simulated, △ ; Measured.

the paddy field was set equal to the water level of 4 cm and irrigation water was supplied at a fixed flow rate, thus excess water overflowed from the outlet. The average vertical and horizontal percolation rates of water during the experimental period were 0.4 and 0.3 cm/day, respectively. For the continuous irrigation method, water was supplied at an average rate of 0.92 m^3/day, which was equivalent to a 2.3 cm/day increase in the water level. Calculated depth of water in the field with the water-holding management practice agreed well with the trend of measured values[7].

For both herbicides the concentrations in paddy water were the same during the first day after the application under both water management conditions. From the second day, herbicide concentrations in the field with continuous irrigation were lower than those in the field with water-holding management. The main reason for this difference was attributed to higher runoff loss of herbicide in the plot with the continuous irrigation. Good agreement between measured and simulated values was obtained for the two herbicides by considering the water management condition and precipitation. Simulated and measured concentrations of herbicides in paddy water and soil are shown in Fig. 3. Runoff losses of herbicides were also calculated by the PADDY-2 model over the experimental period of 32 days. The cumulative runoff losses of molinate and simetryn were 40.5% and 60.4% of the initial applied mass, respectively, under the continuous irrigation management[7]. Meanwhile, the losses under the water-holding management were 1.5% (molinate) and 18.7% (simetryn) due to overflow of paddy water upon significant precipitation events. For reducing the environmental pollution of pesticides due to surface runoff, it is important to regulate paddy water depth using the water-holding management practice.

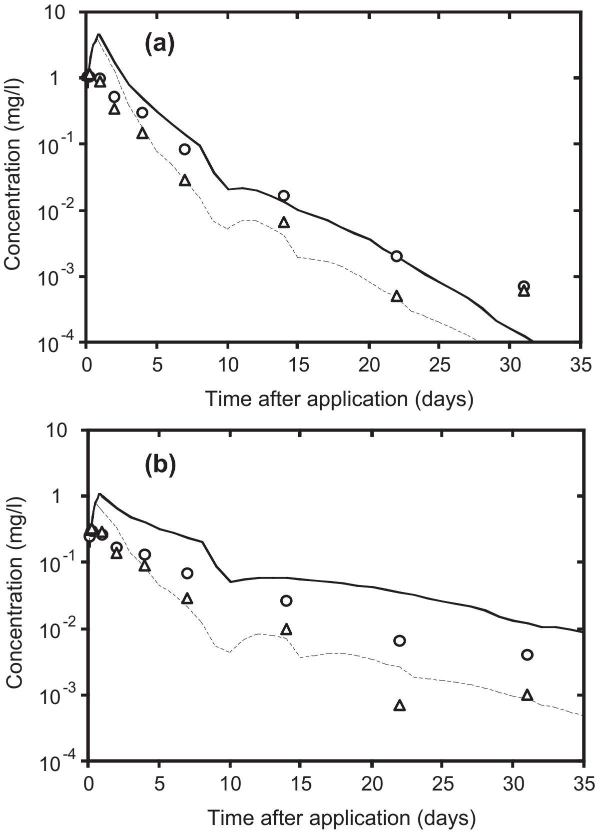

Fig. 3. Comparison between simulated and measured concentrations of herbicides in paddy water[7]

(a): Molinate. (b): Simetryn. Water-holding management: —; Simulated, ○; Measured. Continuous irrigation: ···; Simulated, △; Measured.

3. PADDY-Large model

A landscape-scale simulation model (PADDY-Large), based on the PADDY and PADDY-2 models, was developed for predicting pesticide concentrations in main drainage canals and rivers due to runoff from paddy fields[8]. Depending on the irrigation systems, a rice-producing area was classified into four levels as "individual field plot (30 a)", "farm block comprising twenty field plots and branch canals (6 ha)", "district with a main canal", and "river basin" (Fig. 4). Furthermore, pesticide behavior was estimated focusing on the main canal.

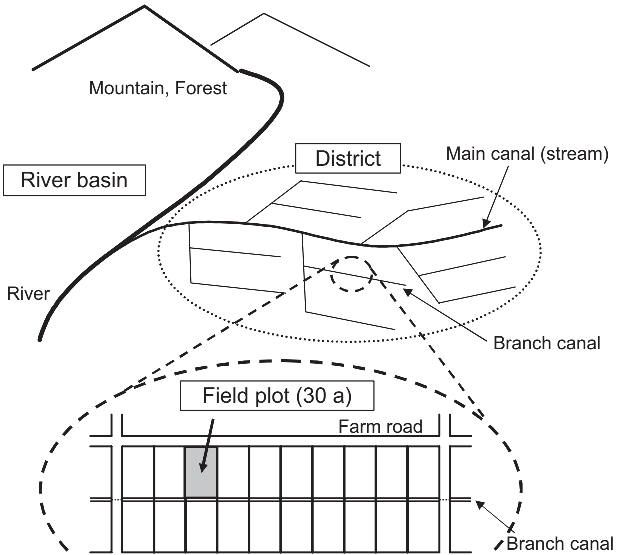

Fig. 4. Hypothetical drainage system for evaluating pesticide behavior in a rice-producing area[8]

Pesticide concentrations in paddy drainage from the field plot can be calculated using PADDY or PADDY-2 models. When pesticides are used as ground applications, generally they are not applied all at once in a farm block. In the model, it is assumed that the distribution of applica-

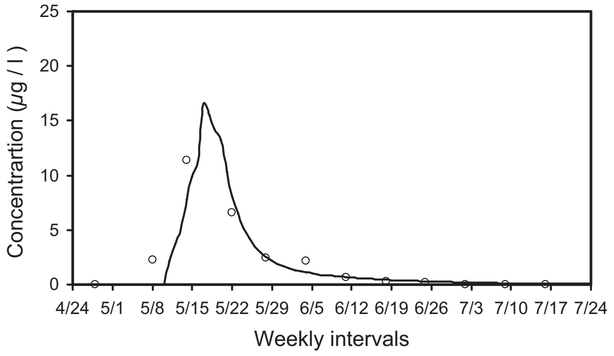

Fig. 5. Simulated and measured mefenacet concentrations in main canal water (1997)[8]
—: Simulated, ○: Measured.

tion dates follows a normal distribution function, and the number of field plots where pesticides are applied at a time in a farm block was estimated by considering the amount of actual pesticide used and the timing of application in a district. For predicting pesticide concentration in a main canal of a district, a continuous stirred-tank reactor model concept was employed[21]. A main canal can be visualized as a series of continuous stirred flow compartments consisting of surface water and sediment solids. The mass balance equations for pesticides in these compartments are expressed in terms of the kinetics of fate and transport processes.

To validate the model, a pesticide monitoring was conducted in a rice-producing area in the southern part of Ibaraki Prefecture in 1996 and 1997. A main drainage canal was located in the center of the catchment area of 271 ha. Paddy fields were spread along the basin of the canal and the total planted area was 55 ha. One-shot herbicides were mainly applied at 5–15 days after rice transplanting (from late April to early May). Herbicide concentrations in the canal increased in early May, reached a maximum in mid May, and declined to below detection limits by early July. Concentrations of mefenacet were higher than those of the other herbicides, because of the wide use of mefenacet covering 61% of the area. Agreement between simulated and measured concentrations of the mefenacet in the main canal was obtained by considering actual pesticide use and environmental conditions in the rice-producing area (Fig. 5).

Simulation of pesticide fate and transport in a paddy field by the PCPF model series

1. PCPF-1 model

The simulation model PCPF-1 has been developed for predicting pesticide concentrations in two compartments, paddy water and 1 cm paddy surface soil layer (PSL)[12,32,33,37]. This model has been used for developing and evaluating field management practices such as irrigation and drainage control and soil management in order to minimize pesticide runoff loss[34,35].

Considering the paddy field environment, the model considers the changes in paddy water depth, precipitation, irrigation, drainage (overflow), vertical and horizontal percolation, and evapotranspiration (sum of evaporation and transpiration) for the water balance. For the chemical mass balance, chemical processes such as dissolution from granules, desorption from the PSL, volatilization, photolysis and the microbial degradation in paddy water and soil are considered. Pesticide transport through the water movement in paddy fields such as runoff loss and leaching is also accounted for. The model program is coded using Visual Basic for Applications in Microsoft Excel®. The input data consist of 23 measured parameters, the daily water balance of the paddy water and local meteorological data. Detailed explanation for the model development and evaluation are given elsewhere[32,33,37].

The PCPF-1 model was evaluated using the results from a field monitoring study with the herbicides pretilachlor and mefenacet which was carried out in the experimental paddy field at NIAES from May 13 to July 15 in 1998. Fig. 6 shows simulated and measured concentrations of mefenacet in paddy water and PSL during the monitoring period[37]. The PCPF-1 model successfully simulated concentrations of mefenacet and pretilachlor in paddy water and PSL with good accuracy in Japanese conditions[33,37] as well as European conditions[12]. PCPF-1 also provides the simulated distribution of herbicide in paddy water and in PSL, and pesticide losses by drainage, percolation and degradation during the monitoring period as shown in the case of mefenacet in Fig. 7[37].

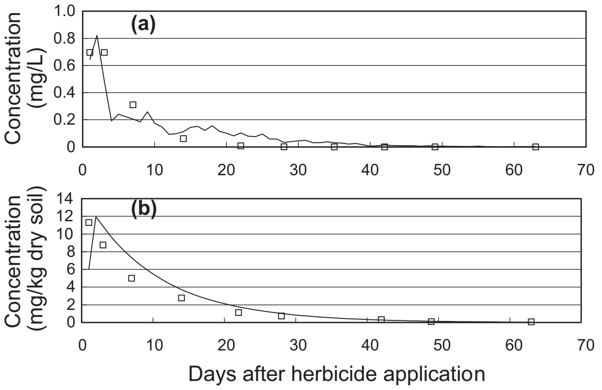

Fig. 6. Simulated and measured concentrations of mefenacet in paddy water (a) and in 0–1 cm surface soil (b) during the monitoring period (1998, Tsukuba, Japan)[37]
□: Measured, —: Simulated.

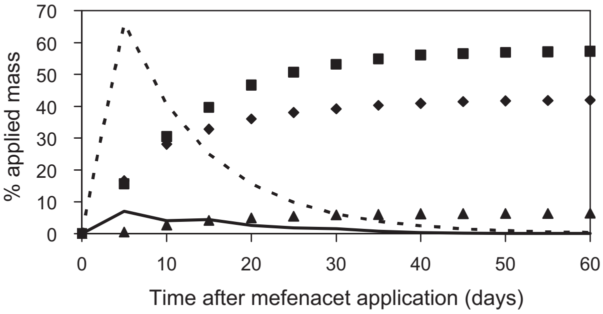

Fig. 7. Simulated distribution of mefenacet in paddy water and paddy surface soil layer, and pesticide loss by drainage, percolation, and degradation during the monitoring period[37]

—: Paddy water, ···: Paddy soil, ◆: Drainage, ▲: Percolation, ■: Degradation.

2. PCPF-SWMS model

The simulation of pesticide transport in paddy soil is performed by the PCPF-SWMS model[28,29] in which PCPF-1 and SWMS-2D models were coupled (Fig. 8). SWMS-2D is an open source FORTRAN coded model in HYDRUS-2D, a Windows-based modeling environment for the analysis of water flow and solute transport in variably saturated porous media[26]. The program solves the Richards' equation for saturated-unsaturated water flow and a Fickian-based advection-dispersion equation for solute transport including a first-order kinetic reaction and the sorption processes by soil/water partitioning coefficient. The model parameters of PCPF-SWMS were calibrated using the observed soil water potentials and chlorine ion tracer in the paddy soil profiles[29].

The coupled model was applied for simulating pretilachlor transport in an experimental paddy plot at NIAES in 1998[33]. The maximum concentrations of pretilachlor in soil water ranged from 0.06 µg/L at 15 cm depth to 0.02 µg/L at 45 and 85 cm depths. In the puddled layer, at 15 cm depth, the pesticide concentration peaked at 29 days after the application. The coupled PCPF-SWMS model can be a beneficial tool to simulate pesticide transport in the soil profile beneath paddy fields[28].

3. PCPF-C model

PCPF-C was developed based on a simplified PCPF-1 model for simulating pesticide fate and transport in a paddy watershed which consists of farm blocks and canals in order to evaluate the best management practices for reducing pesticide runoff[31]. The model simulates pesticide runoff loss from a paddy watershed considering realistic situations of water management and pesticide application procedure in paddy fields. Input data required for PCPF-C consist of parameters for the physicochemical properties of the pesticide, pesticide application procedure, size of the farm blocks and canals, factors for water management in the field such as the water holding period after pesticide application, and the average excess water storage depth during the trial period[31].

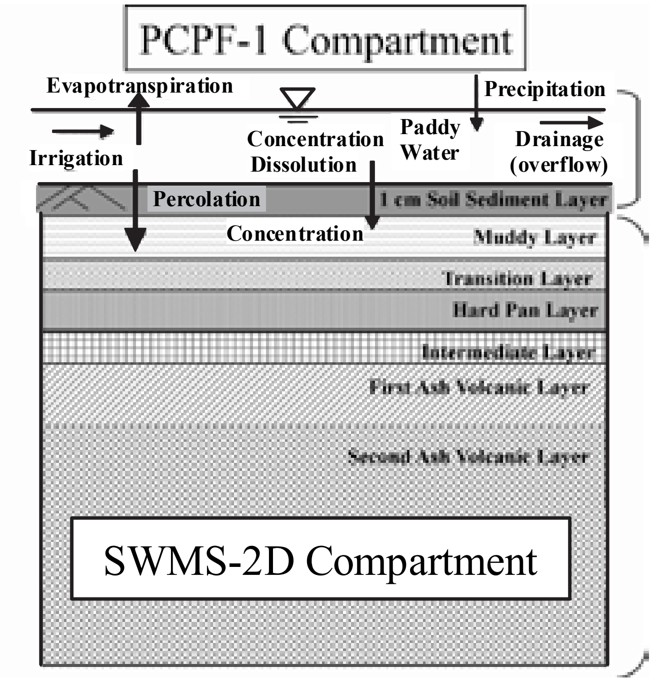

Fig. 8. Conceptual model compartment of the PCPF-SWMS model[29]

The model program which is coded by Visual Basic for Applications in Microsoft Excel® provides the pesticide runoff loss into the canal water through drainage (overflow) and horizontal percolation, and also provides daily pesticide concentration in canal water. For further environmental risk assessment, Monte-Carlo simulation, a widely used method for probabilistic assessment and uncertainty analysis in pesticide fate modeling, is incorporated. Therefore, the PCPF-C simulation can provide a realistic evaluation of the management scenarios for minimizing pesticide runoff from the paddy field to the adjacent surface water systems.

Application of mathematical models to high tier risk assessment in paddy fields in Europe

Although a uniform approach for proper model use has now been established in Europe[4,5], this could not be used in rice due to the unique flooding conditions applied in rice cropping in Europe. Currently, the predicted environmental concentrations (PECs) of pesticides applied in paddy fields are calculated with a Tier 1 spreadsheet which was developed by the Med-Rice group of rice experts[16]. However, the use of more sophisticated mathematical models is essential in cases where refined modeling or application of mitigation strategies have to be applied. The Med-Rice group concluded that, for higher tier risk assessment in rice, the RICEWQ model is the most appropriate model for assessment of exposure in neighboring surface waters. They also pointed out that none of the regulatory models is currently available for calculating PECs in groundwater considering the flooded conditions of paddy fields.

The RICEWQ has been developed in the USA for pesticide registration purpose[40]. This model has been validated under specific scenarios in northern Italy and simulated well runoff processes, but it failed to adequately predict the leaching behavior of the herbicide cinosulfuron[1]. The model calculates pesticide dissipation in paddy water, soil and rice, and also calculates runoff loss from the paddy field due to drainage (overflow) to adjacent surface water systems. However, the model does not consider leaching of pesticides to the deeper soil layer. In order to adequately describe both leaching and runoff of pesticides, an improved version of the RICEWQ model (Version 1.6.2) was developed, which is interfaced between RICEWQ and VADOFT models[12]. VADOFT is a vadose zone flow and transport model contained within the Pesticide Root Zone Model (PRZM) developed by the U.S. Environmental Protection Agency[2]. Evaluation of the RICEWQ-VADOFT model against monitoring data from field studies employed in northern Italy and Greece showed that it can be used as an effective tool for exposure assessments in particular conditions such as wetland rice cultivation[10,11,17]. However, RICEWQ version 1.6.2 could not closely simulate the continuous flow-through systems of irrigation and drainage which are common in rice-cultivating areas in Europe. Therefore, RICEWQ version 1.6.4 was developed, which allows irrigation and drainage to occur concurrently and also distinguishes the different degradation processes (hydrolysis, photolysis, microbial degradation) involved in the dissipation of pesticide in paddy water and soil. Validation of the RICEWQ version 1.6.4 under European rice-cultivating conditions revealed that this model could adequately simulate the water management practices in paddy fields which have a very strong impact on the fate and transport of pesticides in the paddy environment[12].

In Europe, rice-cultivating areas are commonly located in large river basins. Monitoring studies conducted in the main rice cultivation areas in Greece[23], Italy[25] and Portugal[3] revealed the presence of relatively high concentrations of rice herbicides in related surface water systems. Therefore, it will be more relevant to consider risk assessment for rice pesticide at basin scale.

Recent studies revealed that RICEWQ could be an over-conservative tool for predicting the environmental concentrations of pesticides in related surface water systems[18]. However, in the same study, the combination of RICEWQ with the river water quality model (RIVWQ) version 2.02 provided a more realistic estimation of environmental concentrations of pesticides in surface water systems associated with treated paddy fields. Daily based pesticide losses and water releases are generated by RICEWQ due to overflow or controlled drainage, which are considered as chemical and water inputs for RIVWQ at selected junction points along the length of the surface water system. RIVWQ version 2.02 simulates the transport and fate of organic chemicals in riverine systems based on the theory of constituent mass balance[41]. The system geometry is represented using a link-mode approach in which the water system is divided into a number of discrete junctions connected by flow channels. Dynamic constituent transport occurs between junctions via links and is a balance between river-driven flows and dispersion processes. Chemical transformation occurs within each node including dilution, volatilization, partitioning between water and sediment, decay in water and sediment, and re-suspension from bed sediments.

Application of simulation models to controlling pesticide runoff and ecological risk assessment

1. Risk management for reducing pesticide runoff losses from paddy fields

Water management is a key practice for controlling pesticide runoff from the paddy fields. As indicated previously, pesticide concentration is significant in the earlier period and drainage control in this period is crucial for controlling the pesticide runoff[7,24,34,36,38,39]. As reported in those studies, a continuous irrigation-drainage scheme seems to result in significant pesticide runoff losses (e.g. 38% and 49% of total mefenacet and bensulfuron-methyl losses, respectively[36]). Meanwhile, the importance of water management practices such as a water holding period and its practical method for reducing pesticide runoff loss from paddy fields has been discussed[24,30,36,38,39].

The length of the water holding period after pesticide application was evaluated using PCPF-1 simulations for the herbicide mefenacet runoff loss from paddy fields for a continuous irrigation-drainage scheme with water holding periods of 0 day (WH-0), 4 days (WH-4), 10 days (WH-10), 21 days (WH-21), and 30 days (WH-30) as presented in Fig. 9. Obviously, the longer the water holding period was, the smaller the herbicide loss became. In the case of WH 30, i.e. the scenario similar to those in California, the total herbicide loss was minimized up to

3% of applied mass. Therefore, a longer water holding period based on DT_{90} (90% mass dissipation) up to 10 days, for example, is recommended to be a good practice for controlling herbicide runoff[36].

In the Asian monsoon region, a large amount of precipitation is expected during the rice-cultivating season. For reducing pesticide runoff especially in the earlier period after the pesticide application, maintaining excess water storage, which is created by a higher drainage gate in the paddy field to store excess water upon big precipitation events, is a recommended water management practice[24,30,39]. In order to evaluate possible water management practices during the water holding period, prescribed scenarios for a continuous irrigation-drainage scheme and water managements with different excess water storage depths (EWSD) were also compared using PCPF-1 for the herbicide mefenacet runoff loss from paddy fields as shown in Fig. 10. The management practice with a water storage depth of 2 cm gave less herbicide runoff since it could store more water during precipitation events[35].

2. Ecological risk assessment in aquatic environment

In 2005, the Ministry of the Environment of Japan imposed a new pesticide registration scheme concerning ecological risk in the aquatic environment employing the same techniques adopted in the EU and the USA. The evaluation is performed by comparing the acute effect concentration (AEC) for the assessment species with the

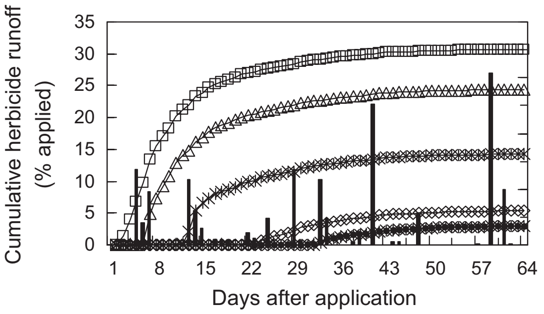

Fig. 9. Simulated runoff losses of herbicide mefenacet responding to different water holding periods (WH) in the paddy fields[35]

▬: Precipitation, -□-: WH-0, -△-: WH-4, -✳-: WH-10, -◇-: WH-21, -✳-: WH-30.

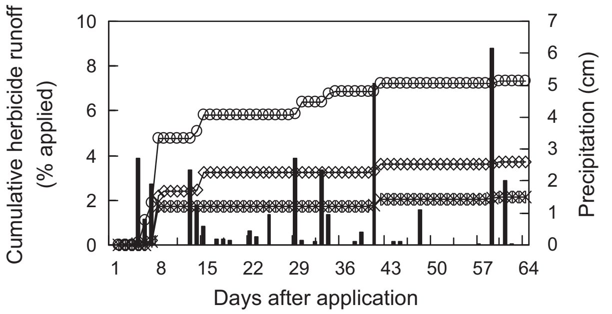

Fig. 10. Simulated runoff losses of herbicide mefenacet by different excess water storage depth (EWSD) management[35]

▬: Precipitation, -✳-: EWSD = 2 cm, -◇-: EWSD = 1 cm, -○-: EWSD = 0 cm.

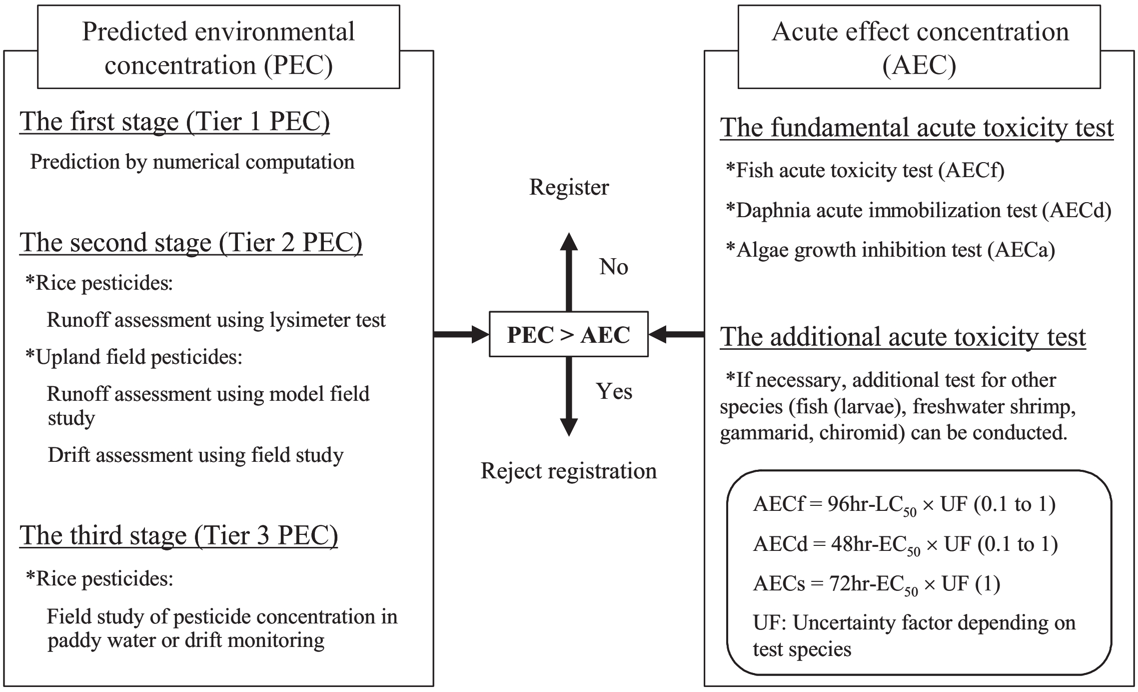

Fig. 11. General scheme for the new Japanese pesticide registration (Modified from Ref. 19)

predicted environmental concentration (PEC) calculated using environmental models and standard scenario for the pesticide concerned (Fig. 11)[19]. The determination of PECs for rice pesticides in river water comprises three tiers[14]: the first tier of preliminary calculation by numerical computation, the second tier using lysimeter tests, and third tier using plot-scale experiments for assessing pesticide runoff and drift[13]. The selected assessment organisms are three typical species: fishes (cyprinodont or carp), crustacean (daphnia), and algae (green algae), and the AECs of the organisms are evaluated from LC_{50} (median lethal concentration) or EC_{50} (median effective concentration) with uncertainty factor depending on test species.

In the EU and the USA, many mathematical models have been evaluated for the pesticide exposure assessment on the regulation and registration process as mentioned above. Although some models for predicting pesticide behavior in paddy fields have been developed under the specific conditions of rice cultivation in Japan, few attempts have been made to evaluate these models for regulatory use. In order to establish a realistic assessment procedure for environmentally-friendly rice production, it is important to develop and validate mathematical models adapted to paddy environments in the Asian region.

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