

Development and validation of a basin scale model PCPF-1@SWAT for simulating fate and transport of rice pesticides



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SUMMARY

The objective of this study was to develop, verify, and validate a new GIS-based model for simulating the fate and transport of rice pesticides in river basins. A plot scale model simulating pesticide fate and transport in rice paddies (PCPF-1) was incorporated into the Soil and Water Assessment Tool (SWAT) basin scale water and pollutant transport model. The new combined model, PCPF-1@SWAT model, was first used on some base-case scenarios to verify that the PCPF-1 algorithm and the routing of variables were correctly implemented. Next, the PCPF-1@SWAT model was calibrated and validated on the Sakura River basin (Ibaraki prefecture, Japan) using mefenacet concentrations measured during the rice growing season in 2008. The modeling procedures for simulating pesticide fate and transport in a Japanese river basin were demonstrated by providing model parameters related to hydrology, land use, pesticide fate, and rice field managements methods.

The water flows predicted by the PCPF-1@SWAT model in the Sakura River basin were accurate throughout the whole simulation year, with R^2 and E_{NS} statistics exceeding 0.74 and 0.71, respectively for daily flow. The use of different seepage rates had appreciable influence on the simulations. High seepage rates gave a slight overestimation of the predicted base flow during the rice growing period, whereas the base flow predictions using lower seepage rates were comparable to measured data.

The PCPF-1@SWAT model successfully simulated the fate and transport of mefenacet in the Sakura River in which measured mefenacet concentrations peaked soon after the initial herbicide application in May, and decreased gradually during the months of June and July. Occasional major precipitation events caused the mefenacet concentration in streams to peak quickly due to a corresponding loss of mefenacet from paddy areas, and then rapidly decrease due to dilution by excess rainfall discharge. The simulation using a seepage rate of 0.12 cm day^{-1} had the most accurate prediction of mefenacet concentration in river water with an R^2 of 0.61 and an E_{NS} of 0.65.

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1. Introduction

In Japan, agricultural land comprise 12% of overall land use (MAFF, 2014). Water quality impact associated with pesticide discharge from paddy fields is a major concern as rice paddies account for about 54% of the total agricultural land (Inao et al., 2008). Indeed, monitoring of Japan's main river systems has revealed the presence of several herbicides commonly used in rice production (Inoue, 1999; Iwafune et al., 2012). The assessment of

the environmental impact of pesticides at a localized and regional scale is a key component for achieving sustainable agriculture. However, the continuous screening of water quality through monitoring and field experiments is not really feasible, and modeling is often the only viable method.

Simulation models can be used as screening tools to prioritize pesticide monitoring efforts and to evaluate the best management practices in controlling pesticide discharges from paddy fields. The PADDY and PCPF-1 models developed in Japan are often used to predict pesticide concentrations in paddy water and paddy soil (Inao and Kitamura, 1999; Inao et al., 2009; Watanabe and Takagi, 2000a,b). Both models were initially developed for

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estimating pesticide concentrations at a field scale. The PADDY-Large model was later developed and validated on a rice producing area in the southern part of Ibaraki prefecture, Japan (Inao et al., 2003). PADDY-Large was also enhanced through the coupling of geospatial information about watershed properties to improve the precision of the model (Iwasaki et al., 2012). Meanwhile, the PCPF-1 model was also recently modified to estimate pesticide concentrations on a larger scale (Phong et al., 2011). However, the scales of the simulated areas of both enhanced models were still limited to a few paddy blocks. Moreover, these models exclusively simulate paddy hydrology and ignore other types of land use which may significantly influence the hydrologic dynamics of river basins. Consequently, the current models cannot adequately simulate the fate and transport of pesticides applied to paddy fields located in river basins consisting of a mix of rice paddies and other types of land use.

The Soil and Water Assessment Tool, SWAT model, is a semi-distributed basin-scale model developed primarily to assist water resource managers in predicting the impacts of management options on water supplies, sediment yields and agricultural chemical transport (Arnold et al., 1998; Srinivasan et al., 1998). The SWAT model has been used extensively across the US, Canada and European countries-areas for which the model is particularly optimized (Coffey et al., 2010). The SWAT model has also been extensively applied in Asia, with successful results reported for many watershed scales and environmental conditions (Ashraf Vaghefi et al., 2014; Park et al., 2013; Somura et al., 2009; Sun et al., 2013; Tang et al., 2013; Tuppad et al., 2011). However, inaccurate results have been reported for some Asian applications, especially when dealing with heavily rice-cultivated areas such as reported by Reshmidevi et al. (2008), Xie and Cui (2011), and Sakaguchi et al. (2014). Thus, some users were constrained to modify the model to improve prediction accuracy under Asian conditions (Im et al., 2007; Kang et al., 2006; Kim et al., 2003; Sakaguchi et al., 2014; Xie and Cui, 2011). The term prediction is, in this study, used as a synonym of hindcasting as defined by Beven and Young (2013). The modifications mainly involved improvements in paddy water balance calculations for the accurate prediction of rice yield, water runoff, or sediment discharge. However, no study involving SWAT has considered the fate of pesticides in rice paddies or pesticide transport from rice paddies to the aquatic environment in river basins.

The main aim of this study is to validate the use of a simulation model for the fate and transport of rice pesticides in river basins. For this purpose, the PCPF-1 model mentioned above was plugged into the SWAT model with the following specific objectives: (i) modification of the current SWAT algorithm for rice paddies, (ii) implementation of the PCPF-1 model into the SWAT model, (iii), verification of the new PCPF-1@SWAT model on a base-case scenario, and (iv) calibration and validation of the PCPF-1@SWAT model using a rice scenario on the Sakura River basin (Ibaraki prefecture, Japan).

2. Materials and methods

2.1. Soil and Water Assessment Tool (SWAT) model

2.1.1. General description

The SWAT model, developed by the Agricultural Research Service at USDA (Arnold et al., 1998), is used to predict the impacts of crop management practices on water including simulation of the hydrologic cycle, plant development, field management practices as well as the transport of sediment, nitrate and pesticides at a river basin scale level. SWAT is able to simulate a single watershed or a system of watersheds by dividing a main basin into several subbasins based on the number of tributaries of the main

river. The size and number of subbasins is variable, depending on the stream network and size of the entire watershed. The subbasins are usually further subdivided into hydrologic response units (HRUs) which correspond to a set of homogeneous combinations of land use, soil and management features (Neitsch et al., 2011a; Xie and Cui, 2011).

The runoff hydrographs are computed based on discharges which are calculated separately at each subbasin and routed through the main river tributaries. The volume of surface runoff from a land surface is estimated with a modified version of the Soil Conservation Service (SCS) curve number method (Boughton, 1989). Sediment transport and consequent chemical transport are simulated based on the Modified Universal Soil Loss Equation (MUSLE; Williams and Berndt, 1977) and Bagnold's equation (Bagnold, 1977). Detailed descriptions of the mathematical processes, parameter relationships and parameter interactions are given in Neitsch et al. (2011b). In addition, the SWAT model was interfaced with the ArcGIS software which provides automated data entry, communication and editing between the Geographic Information System (GIS) and the hydrologic model (Pandey et al., 2005). This hybrid application is known as ArcGIS SWAT (ArcSWAT) and greatly simplifies pre- and post-processing of spatially distributed input data (Neitsch et al., 2011b; Olivera et al., 2006).

2.1.2. Pothole modifications on SWAT

Simulations of paddy fields in the standard SWAT model are performed using the pothole algorithm, which was designed for simulating deep closed depression areas which are hydrologically similar to ponded areas (Neitsch et al., 2011b). The most recent version of SWAT can simulate multiple potholes per subbasin (Beeson et al., 2014) whereas the version of SWAT used in this study (SWAT 2009 rev466) is limited to a maximum of one pothole declared per subbasin. The HRU declared as a pothole can be set for two conditions: ponding or non-ponding. When the pothole is ponding, a water balance algorithm is used to predict the daily fluctuation of water in the pothole due to precipitation, irrigation, outflow and evaporation from the water body. Under non-ponding condition, the SCS curve number method is used to estimate surface runoff. Since potholes are characterized with a conical shape, the volume of precipitation falling into the pothole depends on the surface area of the water body as well as the precipitation rate. When only a portion of an HRU is defined as a pothole, the field runoff generated outside the pothole area will flow to the lowest portion of the pothole rather than contributing to the flow of the main river (Neitsch et al., 2011b).

However, these conventional pothole algorithms were reported to be more appropriate for general closed depressional areas rather than real-world paddy fields (Xie and Cui, 2011). Consequently, the simulated areas under rice cultivation often underestimate water discharge to the main river (Kang et al., 2006; Kim et al., 2003). Therefore some assumptions and algorithms for the use of potholes for rice paddies need to be revised.

As paddies are characterized by a large number of plots separated by low embankments that retain water on the soil surface (Sakthivadivel, 1997), the assumption of conical shape is not realistic. It is reasonable to assume the paddy field to be a shallow box type basin having a constant area in depth, which does not depend on the volume of water stored in the pothole (Xie and Cui, 2011). Therefore, the area of the pothole can be calculated by:

$$SA = A_{HRU} \quad (1)$$

where SA is the total surface area of the rice field (ha) and A_{HRU} is the surface area of the HRU which is assigned to be a pothole (ha).

The daily water balance calculation used in the pothole with ponding water is similar to the water balance considered in

standard models for rice pesticides such as PADDY and PCPF-1 (Inao and Kitamura, 1999; Watanabe and Takagi, 2000b). However, the standard pothole algorithm is still missing the specific characteristics of rice fields such as seepage. The equation used to predict the water level in rice paddy field was therefore modified, by including a seepage term as follows:

$$h_{pw_i} = h_{pw_{i-1}} + RAIN_i + IRR_i - OVER_i - PERC_i - SEEP_i - ET_i \quad (2)$$

where h_{pw_i} is the depth of water in the field at day i (cm), $RAIN_i$ is the daily precipitation (cm), IRR_i is the daily irrigation depth (cm), $OVER_i$ is the depth of overflow discharge from paddies through the drainage/check gate (cm), $PERC_i$ is the daily percolation (cm), $SEEP_i$ is the daily seepage (cm), and ET_i is the daily evapotranspiration (cm). The subscript i denotes the current day while subscript $i-1$ denotes the previous day.

In Japan, the irrigation and drainage schemes which control the ponding depth are usually implemented in order to save irrigation water and to ensure considerable crop yield (Anbumozhi et al., 1998). To accurately simulate rice cultivation, a technique introduced by Guo (1997) which involves three critical depths was implemented in SWAT. The three critical depths are the minimum ponding depth, the optimum ponding depth and the maximum ponding depth. This method has been previously implemented into SWAT with great success (Xie and Cui, 2011). The method was adapted to the water balance algorithms in the PCPF-1 model implemented in SWAT for the irrigation water depth as follows:

$$IRR_i = \begin{cases} h_{norm} - h_{pw_i}^* & \text{if } h_{pw_i}^* < h_{min} \\ 0 & \text{if } h_{pw_i}^* \geq h_{min} \end{cases} \quad (3)$$

$$h_{pw_i}^* = h_{pw_{i-1}} + RAIN_i - OVER_i - PERC_i - SEEP_i - ET_i \quad (4)$$

where h_{norm} and h_{min} are the optimum and minimum ponding depth respectively (cm), IRR_i is the irrigation water depth for day i (cm), and $h_{pw_i}^*$ is the paddy water depth before irrigation (cm). Similarly, the overflow depth is calculated as:

$$OVER_i = \begin{cases} (h_{pw_{i-1}} + RAIN_i) - h_{max} & \text{if } h_{pw_{i-1}} + RAIN_i > h_{max} \\ 0 & \text{if } h_{pw_{i-1}} + RAIN_i \leq h_{max} \end{cases} \quad (5)$$

where h_{max} is the maximum ponding depth (cm), and all other parameters have been previously defined. The total amount of water loss from the paddy field, originating from both overflow and seepage, is referred as paddy discharge in this study.

The three critical depths previously defined can be changed during the simulation since the water ponding depth is usually adjusted throughout the rice growing season in order to achieve highest rice yield (Xie and Cui, 2011). Lastly, the percolation of water from paddy fields occurs under saturated conditions, and it is governed by soil type, ponding depth and paddy soil operation such as puddling, which destroys soil structure and drastically decreases the percolation rate (Tournabize et al., 2006).

Therefore, assigning the three critical depths as well as an average daily percolation rate for the paddy field leads to a more realistic and accurate simulation for the case of rice paddy fields because such data can be readily obtained from the field survey or database. In contrast, obtaining parameters for soil moisture routing techniques used in the conventional SWAT algorithm (Kang et al., 2006) may be difficult to obtain for most of Asian rice-producing regions.

2.1.3. Pesticide fate and transport algorithms

The SWAT model uses algorithms from GLEAMS (Ground Water Loading Effects on Agriculture Management Systems) (Leonard et al., 1987) to simulate pesticide fate and transport in river basins. The process is divided into three components: (i) pesticide

processes in land areas and reservoirs, (ii) transport of one pesticide through a stream network, and (iii) in-stream pesticide transformation and partitioning processes (Larose et al., 2007; Neitsch et al., 2011b). Briefly, the pesticide is partitioned into two forms: soluble, and sorbed with sediments. The ratio between the soluble and sorbed pesticide depends on the equilibrium soil partitioning coefficient of the pesticide. The movement of the pesticide is controlled by its solubility, degradation half-life, and adsorption coefficient against soil organic carbon (Neitsch et al., 2011b). SWAT incorporates a storage feature to lag a portion of the water discharge and lateral flow release to the main river for the case of large subbasins. A simple chemical mass balance developed by Chapra (1997) is used to simulate in-stream pesticide transport and transformations, once the pesticide loadings reach a stream network of a river basin. The model assumes a stream segment to be a well-mixed layer of water overlying a homogenous sediment layer. One pesticide can be routed through the stream network in a given simulation (Neitsch et al., 2011b). Pesticide mass in a stream segment is increased through addition of mass by inflow, resuspension and diffusion of pesticide from the sediment layer. The mass is reduced through removal due to outflow, degradation, volatilization and diffusion into the underlying sediment (Neitsch et al., 2011b).

2.2. PCPF-1 model

2.2.1. General description

The PCPF-1 model is a plot scale model which simulates pesticide concentrations in paddy water and the surface paddy soil layer (PSL). The depth of paddy water is variable depending on a water balance which considers daily irrigation, seepage, percolation, overflow and evapotranspiration (Eq. (2)). The depth of the PSL is null at the beginning of a simulation and increases with cumulative percolation until it reaches a maximum depth of 1 cm where it remains constant. The PSL is considered to be aerobic, so that pesticide degradation occurs under oxidative conditions (Takagi et al., 1998). Both compartments are assumed to be completely mixed reactors having uniform and unsteady chemical concentrations.

In paddy water, the PCPF-1 model considers pesticide fate and transport such as dissolution of the pesticide, pesticide transfer by desorption from the PSL, dilution, concentration, and dissipation by biochemical and photochemical degradation. In the PSL, processes such as equilibrium partitioning of a pesticide between solid and aqueous phases, dissolution of the pesticide, pesticide transfer by desorption from the PSL, percolation and biochemical dissipation are considered (Watanabe and Takagi, 2000b; Watanabe et al., 2006b).

The governing equation for the pesticide mass balance in paddy water is given by Eq. (6):

$$\begin{aligned} \frac{dC_{pw}}{dt} = & k_{DISS}(C_{SLB} - C_{pw}) + \frac{1}{h_{pw}} \left[C_{pw} \frac{dh_{pw}}{dt} \right]_{DISS} \\ & + \frac{1}{h_{pw}} d_{PSL} \rho_{b-PSL} k_{DES} C_{S-PSL} + \frac{1}{h_{pw}} IRR_{w-IRR} - \frac{1}{h_{pw}} (OVER \\ & + LSEEP + PERC) C_{pw} - \frac{1}{h_{pw}} k_{L-A} C_{pw} \\ & + (-k_{PHOTO} f_{US-a} (1 - f_{R-abt}) - k_{BIOCHEM-PW}) C_{pw} \\ & - \frac{1}{h_{pw}} \frac{dh_{pw}}{dt} C_{pw} \end{aligned} \quad (6)$$

where C_{pw} is the pesticide concentration in paddy water (mg L^{-1}), h_{pw} is the depth of water in the paddy field (cm), 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 L^{-1}), d_{PSL} is the depth of

the PSL (cm), ρ_{b-PSL} is the bulk density of the PSL (g cm^{-3}), k_{DES} is the first-order rate constant for the pesticide desorption from the PSL (day^{-1}), C_{S-PSL} is the pesticide concentration in the soil for the PSL (mg kg^{-1} dry soil basis), IRR is the rate of irrigation water supply (cm day^{-1}), C_{W-IRR} is the pesticide concentration in irrigation water (mg L^{-1}), k_{L-A} is the pesticide mass transfer coefficient from paddy water to the atmosphere (cm day^{-1}), $OVER$ is the overflow rate (cm day^{-1}), $LSEEP$ is the rate of lateral seepage (cm day^{-1}), $PERC$ is the rate of vertical percolation (cm day^{-1}), k_{PHOTO} is the first-order rate coefficient of photochemical degradation with respect to the cumulative UV-B radiation ($\text{m}^2 \text{kJ}^{-1}$), f_{US} is the fraction of the UV-B radiation over solar radiation below the rice canopy (–), R_{S-a} is the daily solar radiation above the canopy (kJ m^{-2}), f_{R-abt} accounts for the attenuation by plant growth of the sunlight entering the paddy field (–), and $k_{BIOCHEM-PW}$ is the first-order rate constant of biochemical degradation in paddy water (day^{-1}).

The governing equation for the pesticide mass balance in the PSL layer is given by Eq. (7):

$$\begin{aligned} \frac{dC_{S-PSL}}{dt} = & k_{d-PSL} k_{DISS} (C_{SLB} - C_{pw}) + k_{d-PSL} \left[\frac{C_{pw}}{d_{PSL}} \frac{d(d_{PSL})}{dt} \right]_{DISS} \\ & + \frac{k_{d-PSL}}{(\theta_{Sat-PSL} + \rho_{b-PSL} k_{d-PSL})} \frac{1}{d_{PSL}} PERC \left(C_{pw} - \frac{1}{k_{d-PSL}} C_{S-PSL} \right) \\ & - \frac{k_{d-PSL}}{(\theta_{Sat-PSL} + \rho_{b-PSL} k_{d-PSL})} \rho_{b-PSL} k_{BIOCHEM-PSL} C_{S-PSL} \\ & - \frac{k_{d-PSL}}{(\theta_{Sat-PSL} + \rho_{b-PSL} k_{d-PSL})} \rho_{b-PSL} k_{DES} C_{S-PSL} - \frac{C_{S-PSL}}{d_{PSL}} \frac{d(d_{PSL})}{dt} \quad (7) \end{aligned}$$

where $\theta_{Sat-PSL}$ is the volumetric saturated water content of the PSL ($\text{cm}^3 \text{cm}^{-3}$), k_{d-PSL} is the soil adsorption coefficient of the pesticide in the PSL (L kg^{-1}), and $k_{BIOCHEM-PSL}$ is the first-order rate constant of the pesticide biochemical degradation in the PSL (day^{-1}).

The model has been previously validated in Japan with four compounds: mefenacet, pretilachlor, bensulfuron-methyl and imazosulfuron (Takagi et al., 2012; Watanabe and Takagi, 2000a; Watanabe et al., 2006b). The model was also used and validated in Italy using pretilachlor and cinosulfuron (Capri and Karpouzias, 2007). The PCPF-1 has been evaluated regarding its application potential for paddy fields in California (Luo et al., 2011).

2.2.2. Implementation of PCPF-1 into SWAT

A conceptual schematic of the implementation of PCPF-1 model in SWAT (SWAT 2009 rev466) is illustrated in Fig. 1. The PCPF-1 model calculates the pesticide concentration in paddy water and 1 cm thick soil in a pothole (as the original PCPF-1 model). The daily mass of pesticide loss through leaching below 1 cm surface soil and paddy discharge are transferred into SWAT as variables. The original algorithms from SWAT calculate pesticide transport in the soil environment and the algorithms from GLEAMS simulate pesticide discharge from land areas to the stream network (Gassman et al., 2007). Then a mass balance equation from Chapra (1997) calculates the in-stream pesticide processes in SWAT (Fig. 1).

The latest source code of PCPF-1 (Boulange et al., 2012) was modified: (i) to consider multiple pesticide applications in a rice paddy and, (ii) to simulate the behavior of pesticide sorbed on sediment in paddy water. When pesticide is applied multiple times in a pothole, the pesticide applications are lumped together if the application date is the same. Knowing the amount of pesticide applied on a given day and the pesticide application rate, the surface area where pesticide is applied can be calculated. The pothole is then divided into different areas corresponding to the respective time (day) of pesticide applications. Each area has an independent water balance allowing variations in water holding and field management practices. The water losses through water overflow, percolation, and seepage are computed for each area. The fraction of

pesticide in the sorbed phase is calculated as a function of the pesticide's partition coefficient and the suspended solid concentration in the pothole (Neitsch et al., 2011b).

2.3. Model verification

The implementation of the PCPF-1 model into the modified SWAT model was investigated using a base-case scenario for a 1 km² river basin, entirely covered by paddy fields. A single HRU, declared as a pothole, was defined. Mefenacet was applied at a constant application rate 3 times in 0.5, 0.25, and 0.25 km² of the HRU. Pesticide application timings and the water management used for the base-case scenario are arbitrarily set in order to represent general condition of Japanese rice paddy (Sakthivadivel, 1997) as shown in Table 1. The pesticide fate and transport inputs required by the PCPF-1 model were taken from the previous validation of the model (Watanabe et al., 2006b). Pesticide fate and transport parameters in rivers were set to their default values in the SWAT model.

2.4. Model application

The Sakura River basin, located in southern Ibaraki prefecture (36.2333°N, 140.2833°E), Japan, was used for the calibration and validation of the PCPF-1@SWAT model. The river basin encompasses an area of 345 km²; its main stream is the Sakura River (53.4 km long) which flows into Lake Kasumigaura (Fig. 2). The river has been periodically monitored for rice pesticides (Iwafune et al., 2010, 2011, 2012) and the herbicide mefenacet was detected at relatively high concentrations compared with other pesticides (Inao et al., 2003; Iwafune et al., 2010). The PCPF-1 model has already been validated with this particular compound (Watanabe et al., 2006b) and the environmental behavior of mefenacet in rice paddy is well known (Watanabe et al., 2006a, 2007).

The first year of the simulation (2006) was used to initiate the model and no calibration was attempted during this year. The calibration of the water flow was conducted during the second year (2007) of the simulation (Neitsch et al., 2002). No calibration was attempted on the prediction of mefenacet concentrations due to the limitation of data, and the predictions of both water flow and mefenacet concentration by the PCPF-1@SWAT model were evaluated for the year 2008.

2.4.1. Topographical data

The ArcView SWAT (AVSWAT) interface (Di Luzio et al., 2004) was used for extracting model inputs and outputs. The elevation data was obtained from 1:25,000 scale quadrangle sheet data with a 10 m resolution of from the National Land Numerical Information download service (MLIT, 2013). Stream network maps (created in 2008) and the boundary maps for the basin and subbasins (both created in 2009) were also obtained from the MLIT (2013). The data were provided in the Japan Profile for Geographic Information Standards (JPGIS) format which needs to be converted into vector-type GIS format or shape format for the use in SWAT. A Digital National Land Information (DNLI) conversion tool provided by the Ministry of Land, Infrastructure, Transportation and Tourism was used for this purpose (MLIT, 2013).

2.4.2. Soil and land use data

Four different general soil types were identified in the catchment based on a 1:25,000 scale, digital cultivated soil map data for Ibaraki prefecture in 2007 (NIAES, 2012). The lower and upper part of the Sakura River basins was mostly Gray Lowland soils or Gley soils. The remaining areas in the Sakura River basin were mostly composed of wet Andosols.

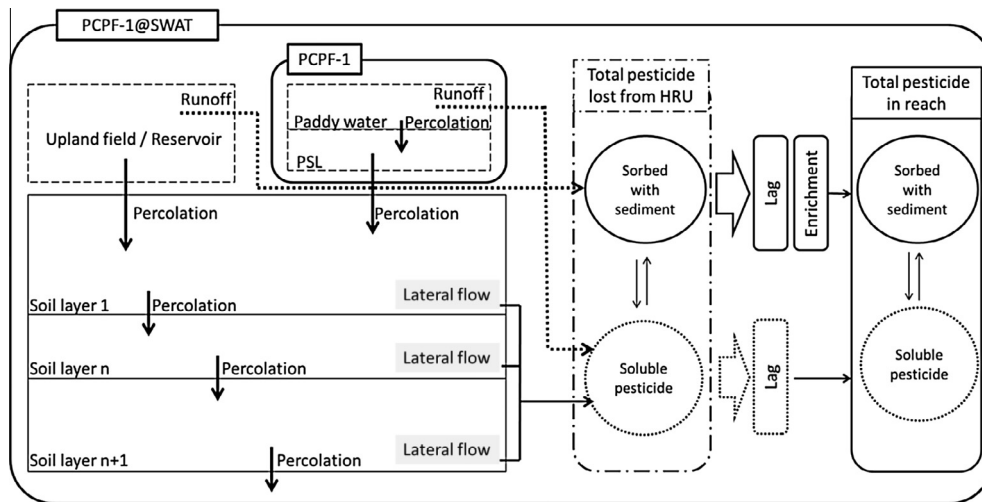


Fig. 1. Implementation of the PCPF-1 model into SWAT.

Table 1

Base-case scenario used for the PCPF-1@SWAT model verification.

Water management scenario of paddy fields	Units	Values
Initial level of water in paddy fields	cm	3.5
Maximum level of water in paddy fields	cm	5
Optimum level of water in paddy fields	cm	3.5
Minimum level of water in paddy fields	cm	1.5
Average daily loss of paddy water to the main channel	cm	0.5
Pesticide scenario		
First pesticide application	days	5
Second pesticide application	days	5
Third pesticide application	days	7
Paddy area of the first pesticide application	%	50
Paddy area of the second pesticide application	%	25
Paddy area of the third pesticide application	%	25
Water holding period	days	5

Note: Day 1 correspond to the start of the simulation. The inputs listed in the pesticide scenario were arbitrary selected to test the PCPF-1@SWAT model.

The land use map of the Sakura River basin used in this study was created in 2008 and was download from the MLIT. The file was available in JPGIS format and was therefore converted to shape format (MLIT, 2013). The river basin was mainly covered with forest, paddy fields and agricultural land which covered 32.5% (112 km²), 27.8% (96 km²) and 17% (58.8 km²) of the entire basin, respectively (Fig. 2).

2.4.3. Climatic data and water flow data

Three years (2006–2008) of daily observed data for precipitation, minimum and maximum temperature, average humidity, and average solar radiation were collected from Radar-AMeDAS-analyzed data base (Japan Meteorological Agency, 2013). Water flow rates at the outlet of the Sakura River basin were acquired for the same period from the observation data of the Water Information System of the Ministry of Land, Infrastructure and Transport, Japan (MLIT, 2013).

2.4.4. Pesticides data and paddy field conditions

Conditions and data regarding mefenacet application were obtained from the study by Iwasaki et al. (2012). Among the 96 km² of paddy fields, about 28.4% were set up as production control to avoid excessive surplus. In these areas, the paddy fields were not used for rice production and were treated as non-ponding paddies in the model. Mefenacet was only applied to 8.1% of the

remaining rice-cropping area. These percentages were estimated by Iwasaki et al. (2012) using the shipment data of mefenacet in Ibaraki prefecture in 1997 and the recommended application rate of 1.05 kg ha⁻¹ of the market product containing mefenacet. The granule formulation of mefenacet is usually applied in paddy fields under flooded conditions 1–2 weeks after transplanting for controlling water grass during the early period of rice cultivation. The mefenacet application dates were determined using the method reported by Iwasaki et al. (2012) where application dates were derived from distribution of rice transplanting date in Ibaraki and the recommended application timing of the market product containing mefenacet.

The physicochemical properties of mefenacet used in the PCPF-1@SWAT are indicated in Table 2. The values were either extracted from the literature or calculated by following SWAT theoretical documentation (Neitsch et al., 2011b). When no guidelines were available, parameters were set to their default values.

The physicochemical properties of mefenacet were assumed to be equal among subbasins and streams. Whereas the equilibrium partitioning coefficient of mefenacet was calculated using the total carbon content of the particular location by the equation below (Wauchope et al., 2002; Weber et al., 2004):

$$k_{d-PSL} = K_{OC} \times \frac{OC}{100} \quad (8)$$

where k_{d-PSL} is the partition equilibrium coefficient of pesticide in paddy surface soil layer (L kg⁻¹), K_{OC} is the sorption coefficient with respect to soil organic carbon of the pesticide (L kg⁻¹) and oc is the total carbon content in the soil (%).

Since the PCPF-1 is, as other pesticide fate and transport models, sensitive to the water balance in the paddy field (Kondo et al., 2012), developing a realistic rice scenario is crucial (Table 3). This scenario was generated to be representative for typical rice practices in Japan (Sakthivadivel, 1997). Since no reliable data regarding water management practices in the river basin were available, these parameters were calibrated to achieve an accurate water flow simulation in 2007. The most influential parameter was the maximum ponding depth, which can be modified throughout the rice season by changing the h_{max} parameter (Eq. (5)). Kondo et al. (2012) showed that parameters related to the calculation of the water balance of rice paddy fields are the most influential when dealing with pesticide fate and transport. The distance between the optimum and the maximum ponding depth is referred to as the excess water storage depth (EWSD), and has been

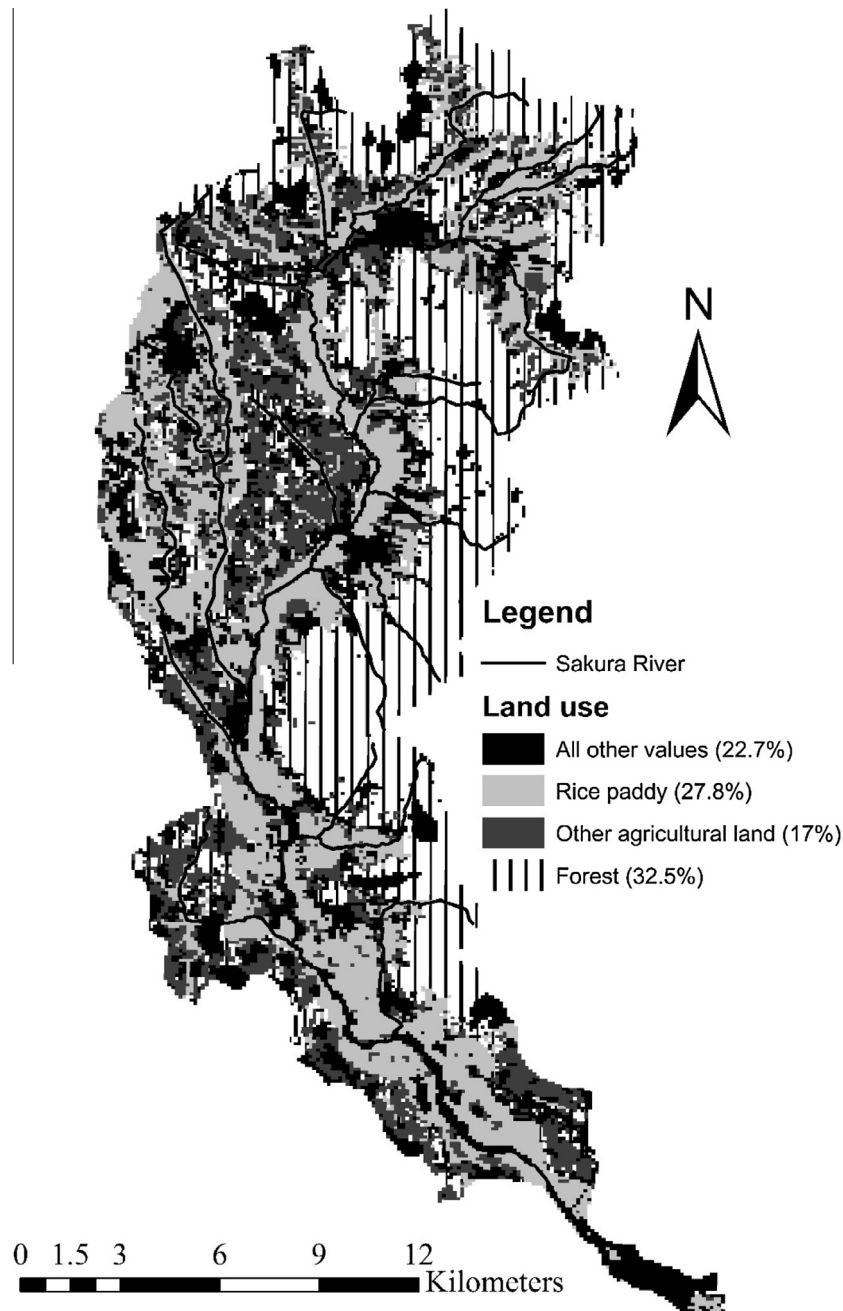


Fig. 2. Sakura River watershed.

reported to be an effective measure to control pesticide discharge from paddy fields (Phong et al., 2008). In general, higher values allow paddy fields to store excess precipitation and are expected to reduce paddy water overflow, which results in lower water flow peaks in rivers.

The water holding period (WHP), also referred to as water holding requirement, is a period during which no water shall be discharged or otherwise spilled from a treated rice field until the specified holding period has elapsed (Linquist et al., 2009). In Japan, the Ministry of Environment requires about 7 days of WHP after pesticide application (JAPR, 2009). This standard WHP was assigned to all paddy fields used for rice cultivation. The WHP will affect paddy-water overflow, however its practice is rather stochastic regarding timing and implementation such as controlling the height of drainage outlet (Kondo et al., 2012).

Typical discharge rates of paddy water into rivers were reported to range from 0.12 to 0.55 cm day⁻¹ (Iwasaki et al., 2012). Consequently, in this scenario, the lateral seepage rate was set for three conditions of 0.12, 0.25, and 0.55 cm day⁻¹ for paddy fields where rice was planted. The daily water loss due to percolation in ponding paddy fields was set to 1.0 cm day⁻¹, a typical value for Japanese paddy fields (Watanabe and Takagi, 2000a).

Puddling assists weed control and homogenization of the soil by destroying aggregates, reduces macropores resulting in a low mechanical strength of the puddled layer which allows easy rice transplanting (Chen and Liu, 2002). However, the efficiency of puddling in reducing percolation depends greatly on soil properties and was proved to be very effective in clay soil while showing reduce effect on coarse soils (Bouman et al., 2007). Since the puddling operation was not available in SWAT, it was replaced by a

Table 2
Input parameters of mefenacet.

Parameters	Units	Values
Water solubility	g mol ⁻¹	5.2 ^a
Soil organic carbon sorption coefficient	ml g ⁻¹	1099 ^a
Dissolution rate constant	day ⁻¹	3.0 × 10 ^{-1a}
Adsorption rate constant	day ⁻¹	4.1 × 10 ^{-2a}
Desorption rate constant	day ⁻¹	4.1 × 10 ^{-2a}
Volatilization rate constant	day ⁻¹	1.8 × 10 ^{-6a}
Degradation rate constant in water	day ⁻¹	3.5 × 10 ^{-2a}
Degradation rate constant in soil	day ⁻¹	3.9 × 10 ^{-3a}
Degradation rate constant in sediment	day ⁻¹	6.9 × 10 ^{-2a}
Sediment–water partitioning coefficient in stream	m ³ g ⁻¹	24.6 ^a
Settling velocity	m day ⁻¹	1.0 × 10 ^{-7b}
Resuspension velocity	m day ⁻¹	2.0 × 10 ^{-3c}
Rate of diffusion or mixing velocity	m day ⁻¹	2.1 × 10 ^{-3b}
Burial velocity	m day ⁻¹	2.0 × 10 ^{-3c}

^a Values taken from Iwasaki et al. (2012).^b Values calculated using the methods described in the Soil and Water Assessment Tool, theoretical documentation (Neitsch et al. (2011b)).^c Default values of the Soil and Water Assessment Tool model (Neitsch et al. (2011b)).

plowing operation. Consequently the percolation rate of rice paddies was not changed after the puddling operation.

2.4.5. Statistical evaluation

Models are approximations of complex processes, their conditional validation shows that their approximation is satisfactory in this limited predictive sense (Young, 2003). For this purpose, the following statistical indices were used to evaluate the prediction accuracy of water flow and mefenacet concentrations: the mean, the standard deviation (STDEV), the coefficient of determination (R^2), the root mean square error (RMSE) and Nash–Sutcliffe Efficiency Index (E_{NS}) (Loague and Green, 1991). R^2 estimates the combined dispersion against the single dispersion of the observed and predicted series. A value of 1 indicates a perfect linear correlation between observed and predicted pesticide concentrations. Higher values indicate less error variance, and values greater than 0.5 are considered typically acceptable (Santhi et al., 2001). A major drawback of R^2 is that since only the dispersion is quantified, a model that systematically over- or under-predicts will still result in good R^2 values (Krause et al., 2005). The RMSE is indicative of the error associated with predictions. The E_{NS} is used to assess the predictive power of hydrological models and indicate how accurately the predicted values match the measured values. It ranges from minus infinity to 1, which is the optimal value (Nash and Sutcliffe, 1970). E_{NS} values greater than 0.5 for monthly flow are considered typically satisfactory whereas values for monthly flow greater than 0.75 are seen as good model performance

(Moriassi et al., 2007). Eqs. (9) and (10) express RMSE and E_{NS} , respectively:

$$RMSE = \sqrt{\frac{\sum (X_{si} - X_{oi})^2}{n}} \quad (9)$$

$$E_{NS} = 1 - \frac{\sum_{i=1}^n (X_{oi} - X_{si})^2}{\sum_{i=1}^n (X_{oi} - \bar{X}_{oi})^2} \quad (10)$$

where $\bar{X}_{oi}$ is the average value of the observed data during the simulation period, X_{si} is the simulated output on day i , and X_{oi} is the observed data on day i .

3. Results and discussion

3.1. Modifications of the pothole module and PCPF-1 implementation

The PCPF-1 algorithm was successfully implemented into SWAT because the predicted mefenacet concentrations in rice fields (paddy water and paddy soil), between the original PCPF-1 and PCPF-1@SWAT models were identical ($R^2 = 1$, $RSME = 0$). Next, the performance of the pesticide multiple applications scheme introduced into the PCPF-1@SWAT model was also verified by comparing the predicted mefenacet concentrations in paddy water and paddy soil in the three areas of the hypothetical watershed. Since the first and second mefenacet applications were scheduled on the same day (Table 1), the water balances and predicted mefenacet concentrations in paddy water and soil in both paddy blocs were identical. Due to the time delay for the third mefenacet application, the predicted mefenacet concentrations in the third paddy bloc had a delayed response with the same concentration range as the first and second mefenacet applications. The cumulative mass of mefenacet lost from each area by paddy water discharge reflected the herbicide loads in each area having different surface areas treated with mefenacet (Table 1).

The summed mass of pesticide lost due to vertical percolation, lateral seepage, and water overflow was equivalent to with the mass of pesticide in the HRU (Fig. 3), indicating no loss of pesticide in the system. Similarly, the mass of pesticide in the river was compared with the mass of pesticide loss through water discharge and lateral flow. Again no evidence of pesticide mass loss in the system was found.

As paddy water is usually clear and undisturbed after the puddling operation, the concentration of suspended solids is usually very low (Valentin et al., 2008) and, therefore, pesticide mass sorbed on suspended solid at a given concentration was low compared with the soluble mass of the pesticide. However, accurate validation on this process remains for the future work.

Table 3
Rice cultivation scenario.

Operations name	Month	Day	Notes/explanations
Plowing	04	15	Land leveling, mix soil layers
Impound	04	20	Start to keep water ponding in paddy fields
Fertilizer application	04	25	Application of basal fertilizers of N:P:K at 40:80:80 kg/ha
Puddling ^a	04	26	Soften the soil for transplanting, mix fertilizer, flatten the soil surface to create uniform soil conditions, control weeds and prevent water leakage
Transplanting	05	01	Transplant the rice young plant into the field
Rice pesticide application ^b	05	01	First rice pesticide application
Mid-summer drainage	07	01	Promote subsurface draining through drying cracks to increase the bearing capacity of the soil for the entry of combine harvester
Harvest and kill	10	01	End of the rice growing season

^a Puddling operation is not available in SWAT and was replaced by a plowing operation which assures a proper mix of soil layers and uniform fertilizer and soil conditions.^b The complete pesticide applications are displayed in Fig. 5a.

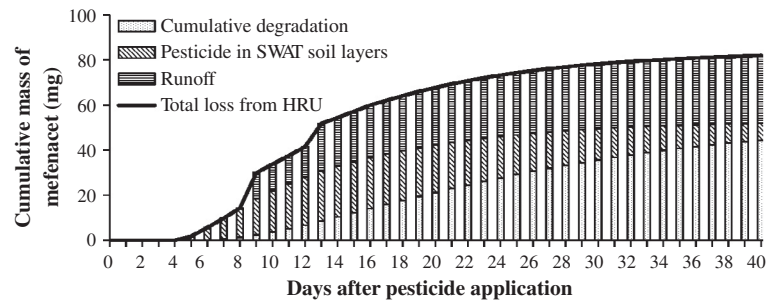


Fig. 3. Comparison of the cumulative mass of mefenacet lost from the pothole and the cumulative mass of mefenacet in the HRU.

3.2. Rice scenario and flow calibration

The modifications of the SWAT pothole algorithm allowed realistic simulations of rice growing areas by capturing the hydrological response of the river basin during the rice growing and non-rice growing (or off-crop) periods. The discharge hydrographs for the calibration and validation periods (2007 and 2008, respectively) are presented in Fig. 4a and b. The validation period is presented only from January through June for the discussion due to the limitation of observed data. The typical rice growing season usually starts from mid-April in Japan (Sakthivadivel, 1997); however, the timing of rice transplanting varies depending on the local agronomical and irrigation schedules. After the rice harvest, rice paddies were kept dry until the next cultivation (Table 3) and were consequently treated as upland fields in the model. As a result, all flow simulations during the off-crop (rice) season were identical. In contrast, from April until rice harvesting at the end of September, ponding water is discharged to the main river due to paddy discharge including seepage (Eq. (2)). Consequently, the observed base flow of the Sakura River basin increased due to the combined effect of paddy and non-paddy discharge caused by frequent precipitation events (Fig. 4a and b). This period

is crucial for the prediction of pesticide concentrations, as they are sensitive to the change in water balance (Wolt et al., 2002). The effect of the different seepage rates on the river flow was clearly observed, and the higher seepage led to the higher base flow of the river. The simulated water flow at the beginning of the rice growing season slightly underestimated the observed data, probably due to the simultaneous ponding of all rice fields in the river basin on April 20 in the simulation scenario (Table 3). The interpretation of management activities across the watershed in the model (field operation timings, pesticide application assumptions) due to the random and irregular drainage operations carried out by different farmers resulted in some minor discrepancies that are unavoidable in river basin modeling (Xie and Cui, 2011). The simulations were nevertheless acceptable for simulating the water flow in the Sakura River basin. The effect of the different scenarios on the prediction of the water flow at the outlet was statistically examined in Table 4. High seepage rate from paddy fields resulted in relatively high mean water flow and RMSE, indicating a general overestimation of the water flow at the basin outlet. Indeed, the RMSE obtained using a daily seepage of 0.55 cm day^{-1} was $16.5 \text{ m}^3 \text{ s}^{-1}$, whereas simulations using daily seepage values of 0.12 and 0.25 cm day^{-1} were more accurate with RMSEs of 4.40 and $2.48 \text{ m}^3 \text{ s}^{-1}$, respectively. The R^2 and E_{NS} were nevertheless satisfactory for all simulations and the lowest values obtained for daily flow throughout the simulations were 0.74 and 0.71 for R^2 and E_{NS} respectively.

3.3. Model validation

The predicted mefenacet concentrations in the paddy fields in the river basin increased after the herbicide application, reaching their maximum within 2 days. The time required to reach the maximum concentration was relatively long compared with other herbicides validated with PCPF-1, but was comparable to the previous validation of the PCPF-1 model using mefenacet (Watanabe et al., 2006b). This pattern may be due to the low solubility of mefenacet (Table 2) and its relatively high application rate (1.05 kg ha^{-1}). In addition, the average k_{d-PSL} value (18 L kg^{-1}) used in the Sakura River basin was significantly lower than the k_{d-PSL} value (24 L kg^{-1}) used during the validation study of the PCPF-1 model, due to the low organic carbon contents in paddy soils in the Sakura River basin. Consequently, mefenacet concentration in paddy water was slightly higher than that of the previous PCPF-1 validation.

Fig. 5a displays the estimated amount of mefenacet applied in the river basin, while Fig. 5b shows the simulated mefenacet concentrations using a WHP of 7 days and daily seepage rates of 0.12 , 0.25 and 0.55 cm day^{-1} . Simulated mefenacet concentrations were sensitive to major rainfall events, which increase the concentration of mefenacet in rivers due to significant paddy field runoff. These concentration peaks in the rivers decline sharply due to water dilution by increased discharge from other crop and non-crop areas.

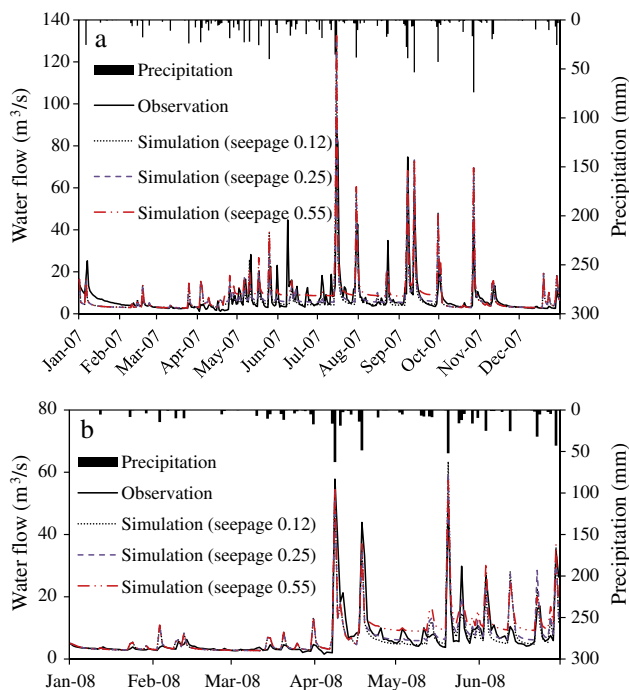
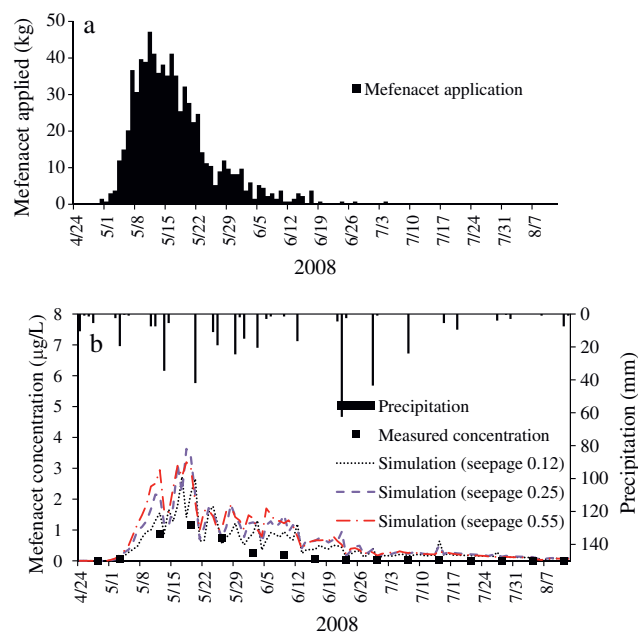


Fig. 4. Observed and simulated water flow at the outlet using a WHP of 7 days and seepage losses of 0.12 , 0.25 and 0.55 cm day^{-1} during (a) the calibration in 2007, and (b) during the validation in 2008.

Table 4

Validation of predicted daily water flow predicted at the Sakura River watershed outlet using a WHP of 7 days with 3 different seepage rates.

Seepage rate (cm day ⁻¹)	Observed (m ³ /s)		Simulated (m ³ /s)		R^2 (–)	RMSE (m ³ /s)	E_{NS} (–)
	Mean	STDEV	Mean	STDEV			
0.12			6.60	7.47	0.74	4.40	0.74
0.25	6.9	8.1	7.11	7.44	0.76	2.48	0.76
0.55			8.15	7.64	0.74	16.5	0.71

STDEV, standard deviation; RMSE, root mean square error; E_{NS} , Nash and Sutcliffe model efficiency.**Fig. 5.** (a) Estimated amount of daily applied mefenacet in the basin, (b) observed and simulated mefenacet concentration at the outlet of Sakura River watershed using a WHP of 7 days and seepage losses of 0.12, 0.25 and 0.55 cm day⁻¹.

The maximum observed mefenacet concentration was 1.17 µg L⁻¹ whereas the simulated maximum mefenacet concentrations were 3.98, 4.37 and 5.73 µg L⁻¹ for seepage rates of 0.12, 0.25 and 0.55 cm day⁻¹, respectively using a WHP of 7 days. Fig. 5b confirmed that the different scenarios greatly affected the predicted mefenacet concentrations at the outlet. High seepage from rice paddies was responsible for high mefenacet concentrations in the rivers.

The R^2 , RMSE and E_{NS} statistics computed for the predicted mefenacet concentrations generated by the three scenarios are presented in Table 5. The statistical indices varied broadly depending on the scenarios. In general, the higher seepage rates were responsible for high RMSE and high mean mefenacet concentrations, which resulted in overestimations of the observed mefenacet concentrations in the river water. The importance of reporting multiple statistics is well illustrated since the R^2 statistic, which only quantify the dispersion, is good for all seepage rates (Krause et al., 2005). Despite the overestimation, given the uncertainty

enclosure into the estimated mefenacet application dates and amounts, the PCPF-1@SWAT model simulated the general trend of mefenacet concentrations. The simulation using the seepage rate of 0.12 cm day⁻¹ was the most accurate and the R^2 and E_{NS} statistics equal to 0.61 and 0.65, respectively, indicated a good level of performance (see Table 5).

The Japanese government has required farmers to comply with a 7 day WHP to facilitate further reduction of the runoff load since 2007. However, farmers' adoption of this WHP implementation had not appeared yet in 2008, since the mefenacet was detected as early as April 27th at the Sakura River outlet (Fig. 5b). This suggest appreciable paddy discharge after the pesticide application (Iwasaki et al., 2012). In addition, a significant amount of paddy water losses may be attributed to lateral seepage as reported in a monitoring study of the river basin having daily seepage values up to 0.22 cm day⁻¹ (Vu et al., 2006).

In order to improve the current simulations, clear and accurate information regarding pesticide use in the river basin is required. More specifically, precise data on mefenacet application dates and applied amounts are needed. Indeed, a sensitivity analysis study using SWAT, which focused on pesticide fate and transport, reported that pesticide application time has much more impact on pesticide fate and transport than either the application rate or the errors originating in the daily rainfall observations (Holvoet et al., 2005). Pesticide use information for pesticide (other than mefenacet) would permit selection of an appropriate starting date and length of WHP in areas where mefenacet was not applied, thus improving the water flow prediction in the river basins.

4. Conclusions

In order to simulate pesticide fate and transport in a river basin, a new model was developed by improving the algorithms related to the hydrology of rice-paddy fields in SWAT, and combining it with the fate and transport model for the rice pesticides, the PCPF-1 model. The newly developed PCPF-1@SWAT model has the ability to simulate pesticide transport from rice paddies to aquatic environments in large watersheds. The algorithms of the model were first verified using base-case scenarios. No error was detected for the pesticide mass balance in the river basin. In addition, mefenacet concentrations predicted by the PCPF-1@SWAT were identical to the original PCPF-1 using the same scenario. The PCPF-1@SWAT model was validated by simulating the paddy field hydrology and the fate and transport of mefenacet in the Sakura River basin (Ibaraki prefecture, Japan). Although missing data

Table 5Validation of predicted mefenacet concentration (µg L⁻¹) at the outlet of the Sakura River watershed in 2008.

Seepage rate (cm day ⁻¹)	Observed (µg/L)		Simulated (µg/L)		R^2 (–)	RMSE (µg/L)	E_{NS} (–)
	Mean	STDEV	Mean	STDEV			
0.12			0.76	0.71	0.61	2.21	0.65
0.25	0.21	0.36	1.08	1.10	0.75	3.46	–9.72
0.55			1.17	1.39	0.84	3.84	–14.7

STDEV, standard deviation; RMSE, root mean square error; E_{NS} , Nash and Sutcliffe model efficiency.

regarding pesticide use (amount used and application timing) and field management practices influence the model performance, PCPF-1@SWAT predicted the observed river flow and mefenacet concentrations in river water with acceptable accuracy. These results are similar to the sensitivity analysis reported by Fohrer et al. (2012) who reported that the key factor for appropriate pesticide fate and transport modeling were the parameterization and spatial distribution of the herbicide application through the watershed.

The new model is able to simulate river basins consisting of a mixture of upland field and rice paddies. Moreover, the paddy fields conditions can be changed from flooded to dry condition allowing continuous annual simulations. The options available to simulate rice paddies are flexible and the model approach can be applied in all rice growing countries. Lastly, the PCPF-1@SWAT model also benefited from the ArcSWAT interface, since the model is easily applicable to other pesticides and chemicals used in an agricultural watershed which includes paddy fields. The model will therefore be useful for ecological risk assessments associated with those pollutants and identifying the critical sites in order to prioritize costly pollutant monitoring studies.

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