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Assessing pesticide fate and transport following modeling approach: A case study of fipronil in the Sakura River watershed, Japan

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Abstract: Modeling approach has considered as an effective alternative method for environmental risk assessment in recent decades. This work aimed to assess the pesticide fate and transport from rice paddy which has higher potential of pesticide runoff compared to upland fields as reported in previous studies. The study area was the Sakura River watershed, Ibaraki Prefecture, Japan. For modeling rice pesticide, the study applied the PCPF–1@SWAT2012 model. The model was used to simulate concentration of a rice pesticide namely fipronil ($C_{12}H_4C_{12}F_6N_4OS$) in 2009. The simulated streamflow and pesticide concentration were calibrated and validated. The results showed that the maximum pesticide concentrations at the monitored point in the wastershed was 0.008 μg/L in rice paddy cultivation season of 2009. In conclusion, the modeling of the pesitcide was successfully performed in the Sakura River watershed by using the PCPF–1@SWAT2012 model. The fate and transport of the pesticide were assessed. Thus, the modeling can be useful tool for environmental risk assessment.

Keywords: The PCPF–1@SWAT2012 model; Pesticide fate and transport; Rice paddy; Rice pesticide; The Sakura River watershed.

1. Introduction

Rice is main daily meals for nearly half of the world's population especially in Asia [1]. The total global rice consumption is increased from 150 million tons in 1961 to 475 million tons in 2016 and predicted continue to rise in the future [1–2]. Maintaining production of rice is very important task for agriculturists. Due to occurrences of various insects, diseases and weeds, rice farmers have been forced to depend on pesticides [3–4]. However, inappropriate use and management of rice pesticides may adversely affect the aquatic environments. Numerous monitoring studies from Europe and Japan have provided evidence that high pesticide concentrations were usually found in rivers during pesticides application periods of rice cultivation season [5–6]. Because pesticides are applied in the rice paddy where rice is cultivated under the submerged condition, pesticide runoff can occur more frequently via drainage or seepage and percolation [7]. Asian countries produce 90% of rice production in the world [2]. As a result, the aquatic environment of these countries may be at high risk of water contamination due to pesticides loss from rice paddy fields.

Japan is the tenth largest producer of rice in the world. Though pesticides use in Japan has decreased, it is still higher compared to other Asian countries [8]. Some studies reported that the loss of pesticides from rice paddies is one of the major non–point sources of pesticide pollution of water in streams or rivers in Japan [7, 9–10]. Sakura River watershed is located about 50 km north–east of Tokyo and one of the popularity monitored watershed. Sakura River watershed is an agricultural watershed with 77.6% of the geographical area under forest and agriculture in Ibaraki prefecture, Japan [11]. During the rice cultivation season of the watershed, pesticides loads of the streams which are elevated due to agricultural drainages from rice paddies, have a potential to cause aquatic toxicities. Recent investigations reported that more than 39 kinds of herbicides, insecticides, and fungicides were detected in the watershed [11–12]. Specifically, in 2007 and 2008, concentrations of herbicides such as bromobutide, daimuron, and imazosulfuron were monitored at more than 2 µg/l in early–mid of rice season while simetryn and bentazone were high in mid and late of rice season. The high concentrations of these pesticides may adversely affect aquatic ecosystems by changing water quality and interrupting the aquatic food chain resulting in the loss abundance aquatic species [13]. Due to these reason, pesticides use in the rice production of Sakura River watershed is of great concern. Therefore, the prediction and assessment of their fate and transport in water is required to minimize the adverse impacts in the aquatic environment of the watershed.

In recent decades, computer models have been developed and widely applied in many fields such as graphics, geology, geography, environment and agriculture. For rice paddy, they have become an advantages management tool since the last two decades. Since a rice paddy model in watershed scale has been required for assessing the potential environmental risks in Sakura River watershed, RICEWQ–RIVWQ, PADDY–Large, and PCPF1@SWAT2012 model could be considered as best candidates (REF). However, the RICEWQ–RIVWQ and PADDY–Large algorithms for runoff and pesticide movement have focused only on simulation of paddy hydrology and ignored other types of land uses, which may significantly influence the hydrologic dynamics and pesticide concentrations of river basins [14]. On the other hand, the PCPF–1@SWAT2012 simulates both hydrologic processes and pesticide transports from the watershed at two phases [15]. First, the upland phase controls the amount of surface runoff and pesticide loadings to the main channel from upland fields. Second, the water or routing phase controls the movement of water and pesticide loadings through the channel networks of the watershed into the outlet. Thus, the PCPF–1@SWAT2012 is a more appropriate model for this specific study. Therefore, this study aims to evaluate a rice pesticide transport at the Sakura River watershed by using the PCPF–1@SWAT2012model.

2. Materials and Methods

2.1. Study area

The Sakura River watershed located in Ibaraki Prefecture, Japan (Figure 1). The watershed area is about 335 km^2 and main stream, namely the Sakura River, which flows into Lake Kasumigaura is 53.4 km long [11, 16]. The topography of the watershed is classified into mountain areas in the north, and flat in the west and southeast of the watershed, with average elevation ranging from 8 to 852 m [17]. The land use in the Sakura watershed consists of forest land (32.0%), rice paddies (28.6%), upland agricultural fields (17.0%), residential land (13.9%), and other land use (8.5%) [11, 18]. With respect to soil types, the lower and upper parts of the watershed are mostly Brown forest, Black, and Gray lowland soils while other parts are mostly composed of Gley and Peat soils [19]. The Sakura watershed generally has a temperate climate; with the average annual rainfall of 1,318 mm. The average daily maximum and minimum temperatures are 19.6°C and 10.1°C, respectively [20].

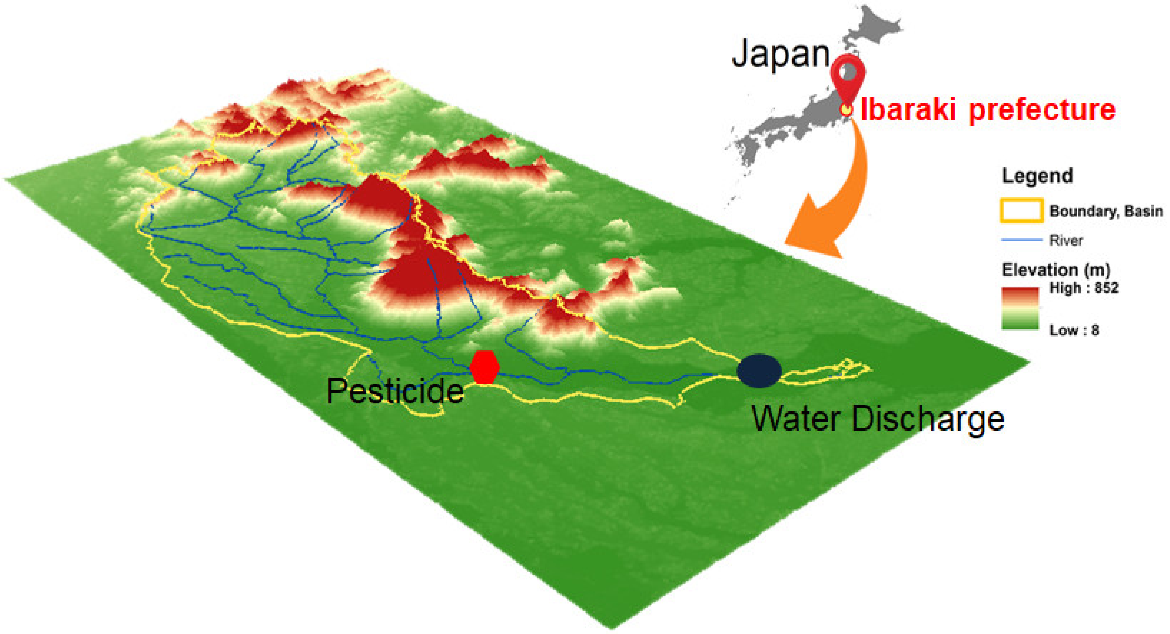

Figure 1. Location and elevation of the Sakura River watershed.

2.2. PCPF–1@SWAT2012 model

2.2.1. Brief model description

PCPF–1@SWAT2012 was updated from the PCPF–1@SWAT which was developed for assessing the impacts of rice pesticides on aquatic environments in watershed scale [21]. Similar to the Soil and Water Assessment Tool (SWAT) model, the PCPF–1@SWAT2012 model also requires topography, land use, soil, weather, crops management practices and pesticide as input data. Figure 2 shows the implementation of Pesticide Concentration in Paddy Field (PCPF–1) model into SWAT model version 2012. In the PCPF–1@SWAT2012 model, rice paddy has been defined as pothole, which is a kind of water bodies for impoundment function in SWAT model. Hence, all performances of the PCPF–1 model are executed inside the pothole of SWAT model. In the PCPF–1@SWAT2012 model, the subbasin can be divided into one or multi hydrologic response units (HRUs). Each subbasin can be set one or multi potholes. When the water is ponding into the pothole, a water balance algorithm is used to calculate the daily amount of runoff. This water balance includes precipitation, water inflow, surface runoff, evapotranspiration, seepage and discharge. In addition, the calculation of water balance components, irrigation process and the pothole variables were redefined. When integrating PCPF–1 model into SWAT model, a procedure to calculate the concentration of pesticide sorbed on sediment was added in the PCPF–1@SWAT2012 model. Because the sediments dissolved in paddy water are not simulated by the PCPF–1 model, pesticide sorbed on soil could not be predicted by overflow [22]. Moreover, recirculation scheme of water was developed. This option aims to calculate the water loss via surface water drainage and tile drainage, which can be collected and re–injected in the field to reduce fresh water requirement.

The PCPF–1@SWAT2012 model was verified and validated in two phases . In the first phase, the pothole algorithms and pesticide mass balance of the model were checked with single and multiple pesticide applications scenarios. The verified results showed that the algorithms used to simulate paddy field water management and pesticide concentrations for single and multiple applications were also correctly implemented into SWAT, and the PCPF–1 was correctly linked to the SWAT model. For the second phase, the

PCPF–1@SWAT2012 model was applied in Sakura River watershed (Ibaraki, Japan) for simulating four herbicide fate and transport. The simulated water flow rate and pesticides concentrations in the Sakura River watershed were good. The model needs to be checked and verified in other watersheds with various pesticides and pollutants.

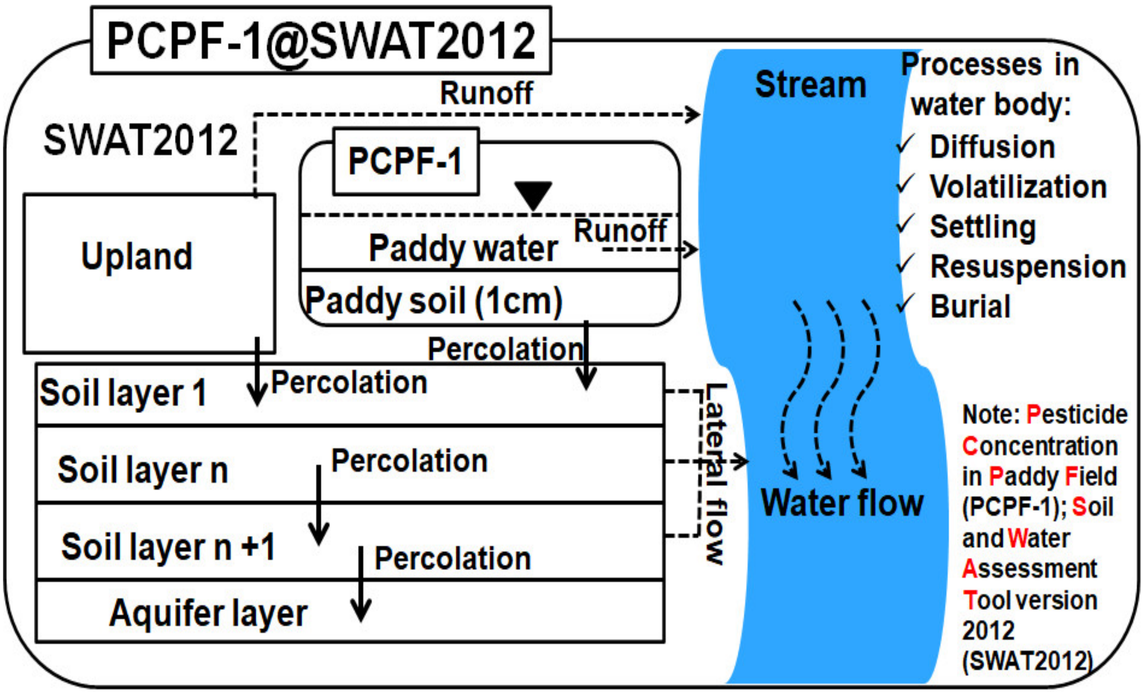

Figure 2. The implementation of PCPF–1 model into SWAT model flowchart [14].

2.2.2. Data collections and processing

The topographic data was obtained from the website of the The Terra Advanced Spaceborne Thermal Emission and Reflection Radiometer Global Digital Elevation Model (ASTER–GDEM) at resolution of 30 m [17]. Stream network and subbasin boundaries data were downloaded from the National Land numerical information download service [18]. The land use data of the Sakura River watershed used in this study were created in 2008 [18]. The data were downloaded from National Land numerical information download service. The dominant land use types of the watershed were forest (32.01%), paddy fields (28.55%), upland agricultural (17.04%), and residential land (13.92%). Paddy fields predominantly covered the west and south parts of the watershed. Soil types were identified in the catchment based on a 1:25,000 digital cultivated soil data for Ibaraki prefecture in 2007 [19]. The dominant soil types of the lower and upper parts of the Sakura river watershed were mostly Brown forest, Black and Gray lowland soils. The remaining parts of the Sakura river watershed were mostly composed of Gley and Peat soils. Two of the above listed data were provided in the Japan Profile for Geographic Information Standards format (JPGIS) under Geographic Projection (JGD 2000), which need conversion into spatial vector–type GIS (shapefile) and SWAT attribute format under Universal Transverse Mercator Projection (JGD 2000 UTM Zone 54). The observed daily data of precipitation, minimum and maximum temperatures were also collected. Four years weather data (2006–2009) were downloaded from Japan Meteorological Agency– Radar–AMeDAS–analyzed data base [20]. Water flow rates at the outlet were acquired for 2008 and 2009 from the observation data of the Water Information System of the Ministry of Land, Infrastructure and transport, Japan [18].

Regarding pesticide data, the PCPF–1@SWAT2012 model requires two groups of pesticide data including application and pesticide properties. The model demands pesticide application time, rate, area, and water holding period (WHP) for creating pesticide input table (Table 1). The application rates of the pesticides were obtained based on shipment amount, usage rate, percentage active ingredient and pesticide product information which were extracted from various literatures especially pesticide database of Japan Plant Protection Association (JPPA) [23]. An insecticide namely fipronil ($C_{12}H_4C_{12}F_6N_4OS$) was selected for the model simulation because the required input data of the pesticides were available.

Table 1. Required pesticides application data for writing the pesticide input table of the model.

Parameter	Unit	Definition	Input file
MGT_OP	none	Operation code. MGT_OP=19 for rice pesticide application	.mgt
MONTH/DAY or HUSC	days	Day and month when the rice pesticide is applied in the HRU	.mgt
pcpfipest	none	Integer that identify the pesticide name	.mgt
pst_pcpfkg	g/m^2	Pesticide application rate	.mgt
pcpfarea	%	Percentage of the HRU where the pesticide has been applied	.mgt
pcpfwhp	days	Water holding period	.mgt

2.2.3. The model evaluation

The study used the Nash–Sutcliffe model efficiency coefficient (NSE), the Root Mean Square Error (RMSE) and Percent bias (PBIAS) to evaluate the prediction performance, tendency and model accuracy [24–27]. NSE can range from $-\infty$ to 1 and an NSE of 1 corresponds to a perfect match between estimation and observations. An NSE of 0 indicates that the model estimations are as accurate as the mean of the observed data, whereas an NSE less than zero ($-\infty$ <NSE < 0) occurs when the model prediction of observed mean is not accurate. Similar to NSE, RMSE is also one criterion most widely used for assessment of model output against observed data. The RMSE values can range from 0 to + ∞ with 0 being a perfect prediction. Because NSE is related normalization of the mean squared error (MSE) and RMSE [28] the PBIAS was additionally calculated. The optical value of PBIAS is 0.0, positive and negative PBIAS values indicate model underestimation and overestimation bias, respectively [26, 29].

3. Results and Discussions

3.1. The model calibration and validation

The Sakura River watershed is divided into 36 subbasins based on hydrological characteristics of the watershed. Simulations have been done for the calibration (2008) and validation (2009) of water discharge in daily time step (Figure 3). Since the rice paddy accounted to 28.5% of the Sakura River watershed area, the water management practices in the paddy field have significant effect on the water flow simulation. However, according to [22] no reliable data regarding that kind of activities in the watershed were available. Therefore, the input data related to the water management practices in the paddy field of the watershed were generated based on assumptions, and those data were extracted from the study [14]. The water management input data include (i) the water holding period, which was 7 days after the pesticides application, (ii) the tile flow rate to channel from paddy fields, which was 0.12 cm/day, and (iii) the percolation rate in the paddy fields, which was 1 cm/day. In addition, the study could only calibrate parameters of pesticide simulation in 2009 due to the observed pesticide data limitation. The selected parameters for calibration of the water discharge and pesticide simulations in the Sakura River watershed are shown in Table 2.

Table 2. The calibrated values of the parameters used for the water flow rate simulation.

Parameter	Description	Unit	Calibrated range	Output variables
GWQMN	Threshold depth of water in the shallow aquifer	mm	0–5000	Water discharge
GW_DELAY	Groundwater delay	day	0–500	
GW_REVAP	Groundwater "revap" coefficient	none	0.02–0.2	
REVAPMN	Threshold depth of water in the shallow aquifer for "revap" to occur	mm	0–500	
LAT_TTIME	Lateral flow travel time	day	0–180	
CN2	Initial SCS CN II value	none	–0.2–0.2	
PERCOP	Pesticide percolation coefficient	none	0–1	Pesticide
CHPST_REA	Pesticide reaction coefficient in reach	1/day	0–0.1	
CHPST_KOC	Pesticide partition coefficient between water and sediment in reach	m^3/g	0–0.1	

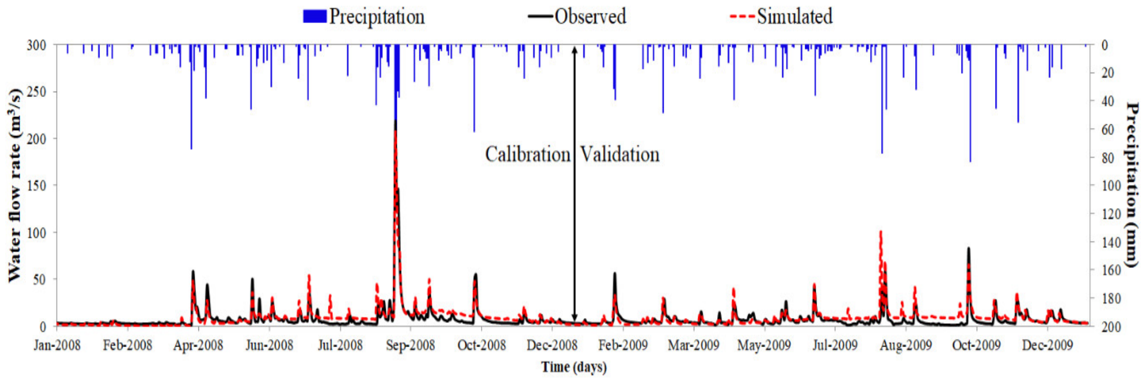

Figure 3. The simulated water flow rate at the outlet of the Sakura River watershed during the 2008–2009 period.

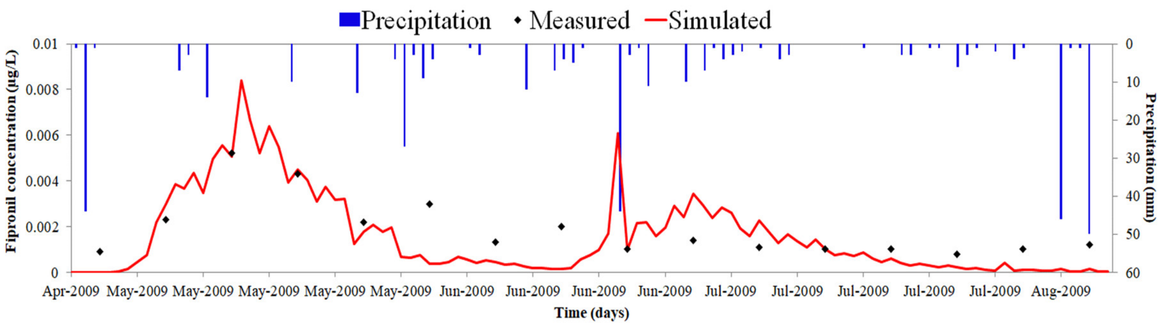

Figure 4. The predicted fipronil concentration for rice season during 2009 at the pesticide monitoring point in the watershed.

Although the simulated baseflow was fluctuated more than that of the observed data, the simulated flow rate showed good response with the observed data and precipitation. Specifically, when the rain came, peak of predicted water flow rate was achieved and vice versa; and the tendency between the predicted flow rate and the observed data was found similar. Table 3 shows the calculated statistical indices for evaluating the model performance of water discharge simulation. The calculated RMSE and PBIAS values, respectively, showed that the predicted water discharge rate had large errors and they were overestimated. Meanwhile, the NSE values indicated that the water discharge rate simulation of the model was good in the calibration year and acceptable in the validation year.

The predicted concentrations of the insecticides were lower compared to the measured insecticides concentrations. The statistical evaluation results of the predicted values of the insecticides concentrations are summarized in Table 3. The NSE and RMSE values were very close to zero. However, the values of PBIAS index indicated that the predicted concentrations of the insecticide in the reach were underestimated possibly because of the insecticide application timing and rate, which did not match with the corresponding actual application timing and rate. In conclusion, the predicted insecticides concentrations were found acceptable.

Table 3. The computed values of model evaluation indices values for the simulated concentrations of the two insecticides.

Variables	Period	NSE	RMSE	PBIAS
Water discharge	Calibration	0.85	6.52	–15.68
	Validation	0.22	7.03	–26.24
Fipronil	Calibration	0.19	0.001	19.12

3.2. Assessment of Fipronil transport

In the Sakura River watershed, the paddy fields are allocated a long with the river in low–land area. So, the applied pesticides in paddy fields are likely to spread to the surrounding aquatic environment. The simulated fipronil concentrations at the monitoring point are displayed in figures 4. The maximum values of the predicted and monitored insecticides concentrations were 0.008 and 0.005 μg/L in 2009 for fipronil. Since insecticides are applied to protect rice against insects throughout the whole growing season, their concentrations in reach increased two times in the 2009 rice season. However, the predicted insecticides concentrations occurred at the beginning of May, rose up in the middle of May, and then decreased.

The pesticides concentrations, which were simulated by the PCPF–1@SWAT2012 model, showed that the rice treated area, application timing, rate and water solubility have strongly affected the prediction. Specifically, the peaks of the insecticides in the middle of May were probably due to nursery–box application upon transplanting. Although fipronil had large rice treated area its peak concentrations of fipronil were low. That can be explained by the application rate of 0.0101 kg/ha, and water solubility values of 3.78 mg/L for fipronil. In addition, the simulated concentrations of the insecticides were low might because of the high Koc (803 ml/g for fipronil) and expected to be mainly applied following the pest forecasting. In other words, the differences in the application rates and methods for the insecticides probably explain why they were detected and simulated at low concentrations. In addition, the pesticide transport was associated with rainfall. When ranfaill eccexed certain amount, it caused loss of the applied pesticide from rice paddy. On the other hand, high rainfall also diluted the pesticide concentration in water bodies.

4. Conclusions

The PCPF–1@SWAT2012 model was applied for predicting transport of a rice insecticide namely fipronil in the Sakura River watershed during 2008–2009 period. The model simulated the observed data with acceptable tendency. The fipronil concentration was increase during rice seasons in the watershed. Strong relationship existed between the increase in the simulated pesticide concentration in the rivers and pesticide application timing and rainfall. However, the model needs to be verified with other pesticides in this watershed as well as in other watersheds. Furthermore, to improve the model accuracy, detailed information regarding water management and pesticide use in the watershed are required.

Author contribution statement: Collected data: L.H.T., H.W.; Analyzed and processed the data: L.H.T.; Model performing: L.H.T.; wrote the draft manuscript: L.H.T., H.W.; Manuscript editing: L.H.T., H.W.

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