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Dynamic modeling of organophosphate pesticide load in surface water in the northern San Joaquin Valley watershed of California

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Major factors governing the instream loads of organophosphate pesticides are magnitude and timing of surface runoff and pesticide application.

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ABSTRACT

The hydrology, sediment, and pesticide transport components of the Soil and Water Assessment Tool (SWAT) were evaluated on the northern San Joaquin Valley watershed of California. The Nash–Sutcliffe coefficients for monthly stream flow and sediment load ranged from 0.49 to 0.99 over the watershed during the study period of 1992–2005. The calibrated SWAT model was applied to simulate fate and transport processes of two organophosphate pesticides of diazinon and chlorpyrifos at watershed scale. The model generated satisfactory predictions of dissolved pesticide loads relative to the monitoring data. The model also showed great success in capturing spatial patterns of dissolved diazinon and chlorpyrifos loads according to the soil properties and landscape morphology over the large agricultural watershed. This study indicated that curve number was the major factor influencing the hydrology while pesticide fate and transport were mainly affected by surface runoff and pesticide application and in the study area.

1. Introduction

Agricultural runoff is the main contributor of nonpoint source (NPS) pollution, which adversely affects surface water and groundwater quality in the United States (Yu et al., 2004; Vazquez-Amabile et al., 2006). As a typical NPS pollutant, pesticide transport and dispersion are driven by the rainfall-runoff process with high spatial and temporal variability in agricultural watersheds. While the application of pesticides has greatly increased the efficiency of farm production, it has also brought about some negative consequences. Variable amounts of pesticides can be released to rivers and aquifers in agricultural watersheds, potentially causing detrimental effects on environment and human health. The San Joaquin Valley, located in the south of the Central Valley of California, is one of the most productive agricultural regions in the world. According to a sampling project in the San Joaquin Valley during 1992–1995, 37% of the stream samples exceeded the pesticide criteria for the protection of freshwater aquatic life (Dubrovsky et al., 1998). Chlorpyrifos and diazinon are of the most commonly used organophosphate pesticides and frequently detected in the surface water and groundwater in the San Joaquin Valley (USGS, 2000; Domagalski and Munday, 2003).

Water quality modeling is emerging as a key component of water quality studies at watershed scale, such as Best Management Practices (BMPs) and Total Maximum Daily Load (TMDL). Modeling serves as a valuable tool in understanding surface water contamination caused by pesticides from agricultural watersheds. The temporal trend in pesticide exposure is commonly addressed by applying data on actual pesticide applications and weather conditions in the model simulations. The spatial variability on pesticide yields is usually incorporated by conducting spatial analyses in Geographic Information Systems (GIS). Reliable hydrological modeling is often the first step in the development of dynamic exposure simulation of pesticide at watershed scale (Novotny, 1994; Lim et al., 2001). In the past years, various hydrologic simulation models have been applied at watershed scales for spatially explicit prediction of hydrologic processes and associated water quality issues. The Soil and Water Assessment Tool (SWAT) is one of those models and is designed to simulate spatially distributed hydrological information under time-varying conditions. The SWAT model has been used extensively in the U.S. and internationally for studying stream flows, sediment yields and nutrient loads (Gassman et al., 2007). There are few but increasing number of studies using SWAT to model watershed-based pesticide processes

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(Du et al., 2006; Kannan et al., 2006; Vazquez-Amabile et al., 2006; Larose et al., 2007).

The dynamic distribution of pesticide residues are not only determined by their applications and physiochemical properties, but also related to spatial heterogeneity of environmental parameters such as weather conditions, landuse and soil properties. By characterizing the spatiotemporal variability in pesticide transport, risks of pesticide pollution can be assessed in a more realistic way compared to a steady-state or homogenous modeling approach (Deksissa et al., 2004; Holvoet et al., 2005). Thus, the general objective of this study was to evaluate the temporal trend and spatial distribution of pesticide loads in surface water of the northern San Joaquin Valley watershed. At first, the SWAT model was calibrated and applied to the field conditions of the study area to simulate the stream flow, sediment and pesticide loads. According to the pesticide use and detection frequency, chlorpyrifos and diazinon were chosen as test agents in calibrating and validating the SWAT model in the northern San Joaquin Valley watershed. Sensitivity analysis was performed to evaluate the propagation of variances in sensitive input parameters for model prediction. By characterizing the temporal trend and spatial variability in pesticide uses and residues to surface water, the results of this study could yield valuable quantitative information on the nature of hydrological and pesticide processes in the northern San Joaquin Valley watershed. Also, results are anticipated to be useful in developing agricultural BMPs in reducing pesticide loads and improving water quality in the agricultural watershed.

2. Methods and materials

2.1. Study area description

The evaluation of the SWAT model was performed in the northern San Joaquin Valley watershed (Fig. 1). The watershed is generally described by the eight-digit hydrologic unit codes (HUCs) of 18040001 (middle San Joaquin and lower Chowchilla watersheds), 18040002 (middle San Joaquin, lower Merced, and lower Stanislaus River watersheds), 18040007 (upper Chowchilla and upper Fresno River watersheds), and 18040014 (Panoche and San Luis Reservoir watersheds). The San Joaquin River at Vernalis, with a monitoring site (#11303500) maintained by the U.S. Geological Survey (USGS), was chosen as the outlet of the simulated watershed since it is the lowest monitoring station on the river not subject to tidal influence (Quinn and Tulloch, 2002). The discharge inlets of the upper San Joaquin, upper Merced, upper Tuolumne and upper Stanislaus Rivers were defined at the USGS monitoring sites of #11251000, #11270900, #11289650, and #11302000, respectively (Fig. 1). The study area includes the majority of agricultural areas in the counties of Stanislaus, Merced, and Madera, and part of San Joaquin and Fresno Counties. The total area is 14 983 km^2, with 9902 km^2 in the San Joaquin Valley, 2182 km^2 in the Coastal Range, and 2899 km^2 in the Sierra Nevada, respectively.

The study area was delineated into 15 subbasins according to natural stream network and irrigation water diversion (Table 1). The subbasin delineation in this study was consistent with the subbasins defined by the California Central Valley Regional Water Quality Control Board (CEPA, 2007a). For each subbasin, multiple hydraulic response units (HRUs) were distributed based on the overlap of landuse and soil features for landscape characterization at finer resolution. A total of 76 HRUs were defined in the study area based on 5% coverage threshold of landuse types in each subbasin. The distributed cropland areas were comparable to those estimated from National Land Cover Data (NLCD) (USGS, 2001) and from landscape surveys by California Department of Water Resources (CADWR). In the study area, landuse types are generally associated with hypsometric levels, e.g., forest and rangeland areas are in high elevation regions and agriculture areas are in the valley floor. Therefore, the landuse-based HRU distribution also indirectly reflected spatial variability in elevation and slope over the watershed.

2.2. Data acquisition

2.2.1. Environmental parameters

Pertinent SWAT input parameter values, such as topography, landscape, and weather conditions, were compiled using databases from various agencies. Data for landscape descriptions, including elevation, landuse, and stream network were obtained from the database of the Better Assessment Science Integrating Point and Nonpoint Sources (BASINS) in which the SWAT model is integrated as a sub-model (USEPA, 2007). Retrieved data included 1:250 000 scale quadrangles of landuse/landcover data, 1:24 000 scale National Elevation Dataset (NED), and 1:100 000

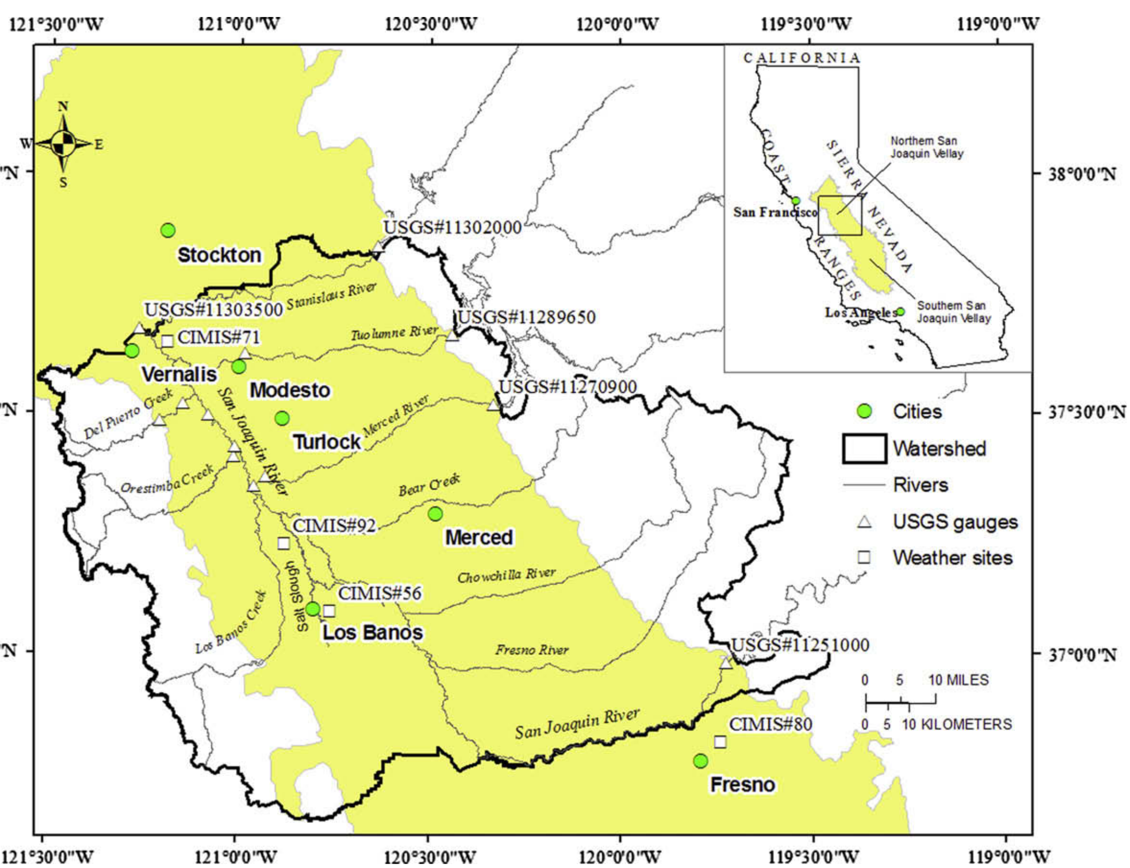

Fig. 1. Study area of the northern San Joaquin Valley watershed.

Table 1
Subbasins delineated for the northern San Joaquin Valley watershed

ID	Name	# Of HRUs	Areas (km^2)	
			Total	Cropland
1	Vernalis North	4	23.4	18.2
2	Stanislaus River	6	569.7	303.6
3	Hospital Creek and Ingram Creek	5	392.3	115.2
4	Tuolumne River	7	992.7	261.1
5	Del Puerto Creek	6	382.4	87.9
6	Northeast Bank	3	317.8	252.1
7	Spanish Grant Drain	4	201.8	101.4
8	Turlock Area	4	459.5	388.0
9	Orestimba Creek	5	563.2	146.2
10	Stevenson	3	113.9	85.4
11	West Grassland Basin	7	1551.1	296.5
12	Merced River	5	834.0	466.9
13	Bear Creek	7	2200.2	649.4
14	Salt Slough	3	2009.7	1075.0
15	Chowchilla & Fresno River	7	4364.3	1809.6

scale National Hydrography Dataset (NHD). The spatial scales of the input data are appropriate for the SWAT simulation at the eight-digit watershed level with subbasin delineation as suggested by the BASINS. Contemporary cropland and irrigation areas in the San Joaquin Valley were defined based on the landuse survey database developed by the California Department of Water Resources during 1996–2004 for the counties of San Joaquin, Stanislaus, Merced, Fresno, and Madera enclosed in the Valley. Cropland information based on the surveys was considered to be representative of agricultural land cover condition of the study area in the reference year of 2002. Soil properties were extracted from the 1:24 000 scale Soil Survey Geographic (SSURGO) database (USDA, 2007) based on soil surveys conducted in the study area during 1990s. Daily weather data, including precipitation, solar radiation, min/max temperatures, relative humidity, and wind speed, were retrieved from the California Irrigation Management Information System (CIMIS). Four weather stations within the study area were used in the SWAT model simulation (Fig. 1).

2.2.2. Fertilizer application

Fertilizer used in the study area was estimated from field survey results and fertilizer sale reports. Based on the cropland areas and average fertilizer application rates for each crop type in the study area (Rauschkolb and Mikkelsen, 1978; Potter et al., 2001), annual total use of nitrogen, phosphorus, and manures were first calculated for each HRU for the reference year of 2002. A previous study indicated that the relative differences between reported fertilizer sales and calculated fertilizer use based on cropland maps was only about 5% for San Joaquin Valley (Potter et al., 2001). Therefore, the annual fertilizer use for each year of the simulation estimated based on the use in 2002 and the trend of reported sales of fertilizing materials on a county basis (CDFA, 2006). Monthly variations in the fertilizer applications were determined based on a survey for major crops in the San Joaquin Valley conducted by King et al. (1998). The survey results were organized as monthly fractions of annual fertilizer use for each crop type. In this study, therefore, monthly fertilizer uses for each crop type were first calculated from the corresponding annual uses. For a HRU, total fertilizer use for a month was determined as the summation of fertilizer uses for all included crop types in the specific month.

2.2.3. Pesticide data

Pesticide applications during 1990–2005 were obtained from the Pesticide Use Reporting (PUR) database maintained by California Department of Pesticide Regulation (CDPR, 2007). California requires 100% reporting of all agricultural, structural, and landscape pesticide use summarized by crop, product, location (geocoded to 1 mi^2), date, and quantity applied. Use amounts of pesticide active ingredients were retrieved from the database as weekly averages for each township/range/section, and distributed into the agricultural HRUs in each subbasin. Pesticide products might be incorporated into the soil for slow release, and it is reasonable to assume a long-term emission of pesticide active ingredients to soil and canopy rather than a pulse input of application (McKone et al., 2007).

Physiochemical properties of diazinon and chlorpyrifos were primarily obtained from the built-in pesticide database in the SWAT model (Table 2). The volatilization transfer coefficient was computed according to the Whitman two-film theory (Ruiz and Gerald, 2001; Neitsch et al., 2005). The pesticide partition coefficient was estimated from the octanol–water partition coefficient (Chapra, 1997). Other mass transport coefficients, such as settling velocity, resuspension velocity, mixing velocity, and burial velocity, were set as their default values suggested by the SWAT model (Neitsch et al., 2005).

2.2.4. Monitoring data

Model initialization and evaluation were based on the monitoring data at selected gauges within the study area (Table 3 and Fig. 1). Data on stream flow and water quality for those gauges were collected from the National Water Information System (USGS, 2007) and California Surface Water Database (CEPA, 2007b). Monthly average stream flow rate was aggregated from daily data. In cases where data did not exist for a given month, the long-term monthly average was applied. Sediment concentrations were usually available in a monthly or biweekly interval. Instantaneous sediment load was calculated as the product of measured sediment concentration and stream flow, and then averaged as monthly data. Continuous monthly sediment loads were required by the SWAT model at the upstream inlets (Fig. 1). Therefore, the ESTIMATOR program developed by the System Analysis Branch of the USGS was used to predict missing sediment loads based on measured stream flow (Helsel and Cohn, 1988; Gilroy et al., 1990; Kratzer and Shelton, 1998).

For the period of 1992–2005, about 200 water samples for diazinon and chlorpyrifos concentrations were collected at the watershed outlet (USGS site #11303500), Merced River outlet (#11373500), and Orestimba Creek outlet (#11274538). For other monitoring sites, less than 50 samples were available. Similar to the estimation of sediment loads, the pesticide load in each sampling day was calculated by the corresponding concentration and stream flow. Monthly load was estimated from average daily loads available in that month. In the San Joaquin River watershed, almost all of the agricultural pesticide application is in the San Joaquin Valley (CDPR, 2007). Therefore, the stream water at the discharge inlets of upper San Joaquin (USGS site #11251000), upper Merced (#11270900), upper Tuolumne (#11289650), and upper Stanislaus (#11302000) Rivers (Fig. 1) was assumed to be free of pesticides.

Table 2
Physiochemical properties and mass transfer coefficients for diazinon and chlorpyrifos

Parameter	Description	Values	
		Diazinon	Chlorpyrifos
SKOC[a]	Soil adsorption coefficient (–)	1000.0	6070.0
WOF[a]	Wash-off fraction (–)	0.9	0.65
HLIFE_F[a]	Half-life on foliage (day)	4.0	3.3
HLIFE_S[a]	Half-life in the soil (day)	40.0	30.0
WSOL[a]	Solubility (mg/L)	60.0	0.4
MW[b]	Molecular weight (g/mol)	304.4	350.6
HENRY[b]	Henry's law constant (–)	3.0×10^{-5}	3.0×10^{-4}
CHPST_REA[b]	Hydrolysis coefficient (day^{-1})	0.005	0.012
SEDPST_REA[b]	Degradation coefficient in sediment (day^{-1})	0.043	0.005
CHPST_VOL[c]	Volatilization coefficient in water (m/day)	0.007	0.060
CHPST_KOC[c]	Partitioning coefficient (m^3/g)	8.9×10^{-5}	0.003

[a] SWAT built-in pesticide property database.
[b] Agricultural Research Service (ARS) pesticide property database (USDA, 2001), data for 20 or 25 °C.
[c] Calculated values.

Table 3
USGS monitoring sites used for model initialization and evaluation in this study

Tributary outlets or river site	USGS ID	Location		Sampling type	
		Latitude	Longitude	Stream flow	Water quality
Sites for inlet discharge (Fig. 1)					
San Joaquin River	11251000	36.98	−119.72	×	×
Merced River	11270900	37.52	−120.33	×	×
Tuolumne River	11289650	36.67	−120.44	×	×
Stanislaus River	11302000	37.85	−120.64	×	×
Sites for model evaluation					
Merced River	11272500	37.37	−120.93	×	
	11273500	37.35	−120.96		×
Orestimba Creek	11274538	37.41	−120.02	×	×
San Joaquin River at Cross Landing	11274550	37.43	−121.01	×	×
San Joaquin River at Patterson	11274570	37.50	−121.08		×
Del Puerto Creek	11274630	37.49	−121.21	×	
	11274653	37.52	−121.15		×
Tuolumne River	11290000	37.63	−120.99	×	×
Stanislaus River	11303000	37.73	−121.11	×	×
San Joaquin River at Vernalis	11303500	37.68	−121.27	×	×

2.3. Simulation Scenario and model evaluation

2.3.1. Model background and simulation design

The SWAT model was developed by the U.S. Department of Agriculture (USDA) (Arnold et al., 1998) to predict the impact of land management practices on water, sediment and chemical transport in large complex river basins over a long period of time. The pesticide component of SWAT simulates pesticide transport in dissolved and particulate phases with surface and subsurface hydrologic processes. The fate and behavior of pesticide are determined by its solubility, degradation half-life, and partitioning coefficients (Neitsch et al., 2005). The SWAT simulator version 2005 and its ArcSWAT interface (Di Luzio et al., 2004; Winchell et al., 2007) were used in this study. The interface is integrated with ESRI ArcGIS GIS pre-processor and uses raster- and vector-based spatial data layers of elevation, soil, landuse, and weather as basic inputs to the model. Initialization of the SWAT model also involved parameter settings for lapses of precipitation and temperature, and background concentration of nutrient in rainfall and groundwater. For those parameters, results from literature reviews (TetraTech, 2003, 2004; NADP, 2007) were applied to replace the default values in the model.

The SWAT model provides multiple options for runoff generation and evapotranspiration estimation. In a preliminary analysis, the best combination of runoff generation and evapotranspiration calculation in the study area was identified as the curve number and Priestley–Taylor method, respectively. Although SWAT is based on daily simulation, it is difficult to accurately capture daily results because of possible time shifts in the precipitation, agricultural activity, and measurements for flow and water quality data. Therefore, model predictions were reported and evaluated on a monthly and annual basis. The SWAT simulation was performed for the period of 1990–2005, which included the first two years as the model initialization period. The initialization period for simulation was applied to allow state variables to be calculated from forcing variables rather than user-defined initial values, which might not reflect actual temporal variations. Calibration of the SWAT model was performed for years of 1992–1997, while years of 1998–2005 were used for model validation.

2.3.2. Sensitivity analysis

Sensitivity analysis was performed to identify the parameters that affected the SWAT model predictions significantly. In a conventional sensitivity analysis, the sensitivity (S_I) of output to changes in input is expressed as:

$$S_I = \frac{\partial P}{\partial I}\frac{I}{P(I)} \tag{1}$$

where I is the model input, and P is the model prediction based on the corresponding input I. Eq. (1) implies that partial derivatives of input parameters are assumed to be constant over the range of the uncertainties. One of the widely used analytical estimates for sensitivity analysis is obtained by using central differences for the derivatives:

$$S_I = \frac{\Delta P}{\Delta I}\frac{I}{P(I)} = \frac{P(I+\Delta I) - P(I-\Delta I)}{2\Delta I}\frac{I}{P(I)} \tag{2}$$

The estimate of sensitivity provides a transfer function to propagate the relative error of the target parameter into the relative error of the prediction. The bigger the absolute value of S_I is, the more sensitive the parameter is for the specific model prediction. A negative sensitivity indicates that the parameter has an inverse effect on the prediction. The parameters selected for the SWAT sensitivity analysis are listed in Table 4. The derivative values may vary with the magnitude of input and output values due to the nonlinearity in the model equations (Luo and Yang, 2007). Therefore, the above equation should only be applied on a calibrated model to evaluate sensitivities on a well-defined hydrologic system. In this study, model predictions from the sensitivity analysis were set as the model outputs of the entire watershed for the whole simulation period of 1992–2005, including the total surface runoff within the watershed and stream flow, sediment and pesticide loads predicted at the watershed outlet.

Table 4
Overall sensitivities of selected parameters in the SWAT model calibrated in the northern San Joaquin Valley watershed during 1992–2005

Input parameter	Surface runoff	Stream flow	Sediment load	Dissolved load	
				Diazinon	Chlorpyrifos
CN2	5.50	0.05	0.75	5.57	7.53
GWQMN	0.00	−0.08	−0.16	−0.05	−0.03
SOL_AWC	−0.45	−0.17	−0.29	−0.15	−0.19
SPCON			1.01	−0.60	−0.46
CH_EROD			0.31	−0.04	−0.10
HLIFE_S				0.37	0.78
SKOC				−0.25	−0.62
HENRY				−0.05	−0.31

CN2 SCS: runoff curve number for antecedent moisture condition II.
GWQMN: threshold depth of water in the shallow aquifer required for base flow to occur (mm). SWAT default value = 0.
SOL_AWC: available water capacity of the soil layer.
SPCON: linear parameter for calculating the maximum amount of sediment that can be reentrained during channel sediment routing. SWAT default value = 0.0001.
CH_EROD: channel erodibility factor. SWAT default value = 0.
PERCOP: pesticide percolation coefficient. SWAT default value = 0.5.
HLIFE_S, SKOC, and HENRY are defined in Table 2.

2.3.3. Model evaluation methods

Descriptive statistical analyses were performed to evaluate whether the frequency distribution of predicted data was similar to the measured one. In addition, model-to-data plots were analyzed for possible trends. The linear relationship between pesticide application and corresponding dissolved loads was evaluated by the Pearson's correlation analysis. The model performance (goodness of fit) was primarily evaluated by the Nash–Sutcliffe (NS) coefficient (Nash and Sutcliffe, 1976) as recommended by the American Society of Civil Engineers (ASCE, 1993). The Nash–Sutcliffe coefficient, also called the coefficient of efficiency, indicates how well the plot of the observed versus simulated data is close to the 1:1 (equal value) line. The coefficient is calculated as:

$$NS = 1 - \frac{\sum_j (O_j - P_j)^2}{\sum_j \left(O_j - \overline{O}\right)^2} \tag{3}$$

where O and P are observed and predicted values, respectively, and j is a running index for elements in the data sets. The Nash–Sutcliffe coefficient ranges from negative infinity to 1. The simulation results were considered to be good if the coefficient was larger than 0.75, and satisfactory if it was between 0.36 and 0.75 (Van Liew and Garbrecht, 2003; Larose et al., 2007). In addition to the Nash–Sutcliffe coefficient, the coefficient of determination (R^2) and the root mean square error (RMSE) were also calculated to provide additional statistical information on the model performances. Significance levels of correlation results were classified according to associated p-values (non-significant correlation with $p > 0.05$, strong correlation with $p < 0.001$, and moderate correlation with p value in between). For some model predictions, such as the sediment and pesticide loads at tributary outlets, there were insufficient corresponding measurements for generating NS, R^2 or RMSE. For those variables, the model performance was evaluated by comparing the averages of predicted and observed data during a given time period. All statistical analyses were done with SAS version 9.1 (SAS, 2004).

2.3.4. Model calibration and validation

The SWAT was calibrated for stream flow, sediment, and pesticide load measured at USGS monitoring gauges located on the San Joaquin River and its major tributaries in the study area (Table 3). The longest-running monitoring gauge at the watershed outlet, USGS #11303500 (San Joaquin River at Vernalis), was selected as the primary location for model calibration and validation. This site receives stream flow from all upstream portions of the study area and thus characterizes the water quality in general. In addition, gauges with shorter periods of record were also used during the model evaluation procedures (Table 3). The use of those tributary subbasins ensured that the model was accurately simulating the hydrologic and chemical transport processes on various spatial scales in the study area.

The most sensitive model parameters were chosen in the calibration procedure based on literature review and preliminary sensitivity analysis results (Table 4). The selected parameters are governing factors in simulating the hydrology and pesticide transport processes at watershed scale. Initial value for curve numbers in condition II (CN2) in each HRU was estimated based on landuse and soil hydrological group via the ArcSWAT interface, whereas the target data range of CN2 was established based on values recommended by the USEPA for various crops in the San Joaquin Valley (USEPA, 2002, 2004). Adjustment of the curve numbers within the pre-established range was made to reflect the crop and surface conditions in the study area. As a result of model calibration, CN2 were reduced by 5% of the original value for agricultural HRUs, indicating that the cropland in the northern San Joaquin Valley had a better soil drainage, in terms of soil, landuse, and agricultural management practices, compared to the default conditions derived from landuse and soil data. In the calibrated SWAT model, CN2 values for agricultural HRUs ranged from 67 to 87 for eastern subbasins and 77 to 87 for western subbasins, respectively. Those values were comparable to the recommended CN2 values by the USEPA. For parameters without field measurements, default values were applied first in the SWAT simulation. Parameter modifications were conducted based on the appropriate ranges as defined in the SWAT model documentations. For example, channel erodibility factor (CH_EROD) was adjusted for each subbasin individually with a value up to 1.0. The linear parameter for sediment transport capacity of channels (SPCON) was increased to 0.0002 to lower the amount of sediment deposition. To generate the best fit between predicted and observed pesticide level in surface water, the partitioning of soluble pesticide between percolation and surface runoff (PERCOP) was decreased to 0.2 for diazinon.

3. Results and discussion

3.1. Model sensitivity

Table 4 shows the sensitivity values of SWAT model predictions on selected input parameters in the study area during 1992–2005. The curve number was the most sensitive parameter for the prediction of surface runoff and pesticide loads. The strong positive effect of curve number on surface runoff prediction, with a sensitivity value of 5.50, was neutralized by its negative effects on lateral and base flows, resulting in a moderate sensitivity value of 0.05 for stream flow. This is physically reasonable because elevated curve numbers would represent increased surface runoff and decreased infiltration. For the instream sediment transport processes, the linear parameter of sediment routing capacity (SPCON) had stronger effects on sediment load, compared to the channel erodibility factor (CH_EROD). The elevated load and concentration of suspended sediment would reduce pesticide in soluble phase by partitioning, resulting in a negative sensitivity for dissolved pesticide loads. Among the physiochemical properties, soil-related parameters such as half-life in soil (HLIFE_S) and soil adsorption coefficient (SKOC) were sensitive parameters in predicting dissolved loads of diazinon and chlorpyrifos. The prediction of dissolved chlorpyrifos load was also sensitive to the volatilization process, indicated by the sensitivity value of the Henry's law constant (HENRY). Other physicochemical properties associated with in-stream pesticide transport processes, such as hydrolysis constant and solubility, had minimal effects on the pesticide load predictions.

Results of sensitivity analysis identified the major transport processes and associated governing parameters in determining pesticide fate and distribution in the study area. With large SKOC values, diazinon and chlorpyrifos have moderate-to-low mobility in the soils and are not usually detected in groundwater. Therefore, surface runoff is an important pathway for pesticide transport towards surface waters. After incorporation with the soils by direct application and wash-off from crop canopy, pesticide inventory available for runoff transport is primarily controlled by degradation process in the soils.

Pesticide water–sediment partitioning, which is also estimated based on SKOC and suspended sediment concentration, is the key process for predicting pesticide transport in channel flows in the study area.

3.2. Model performance for stream flow and sediment

Fig. 2 shows the SWAT-predicted monthly stream flows at the outlet of the watershed compared to measured data by the USGS. The Nash–Sutcliffe coefficient was 0.95, indicating a good simulation of hydrology at the entire watershed level. Also illustrated in Fig. 2 are monthly precipitation totals observed at a representative station for the northern San Joaquin Valley watershed at Modesto, CA. Although water flow in the study area had a large contribution from upstream discharges from Sierra Nevada, the predicted stream flow was significantly correlated to the representative precipitation within the northern San Joaquin Valley watershed ($p < 0.001$).

The statistical results of the model performance for stream flow predicted at selected sampling sites during both calibration and validation periods are summarized in Table 5. The simulation generated good or satisfactory results in the comparison with measured data, especially for the San Joaquin River and the major eastern tributaries of Merced, Tuolumne, and Stanislaus Rivers. SWAT model performance in predicting surface hydrology in large agricultural watershed is related to the understanding and characterization of irrigation water diversion and agricultural drainage. For the Tuolumne and Stanislaus River subbasins, two large irrigation districts (Modesto Irrigation District and Turlock Irrigation District) received irrigation water from upstream storages on the Tuolumne and Stanislaus Rivers (Quinn and Tulloch, 2002). Most of the agricultural drainages in the two irrigation districts were discharged to the San Joaquin River. For the Merced River subbasin, for example, irrigation water is delivered by the Merced Irrigation District from the Merced River within and upstream of the study area, resulting in an appreciable decrease of stream flow in the river. For example, 409.8 million m^3 of water from the Merced River was diverted by the district during 2004 (MID, 2004). Consequently, the 2004 annual average stream flow in Merced River

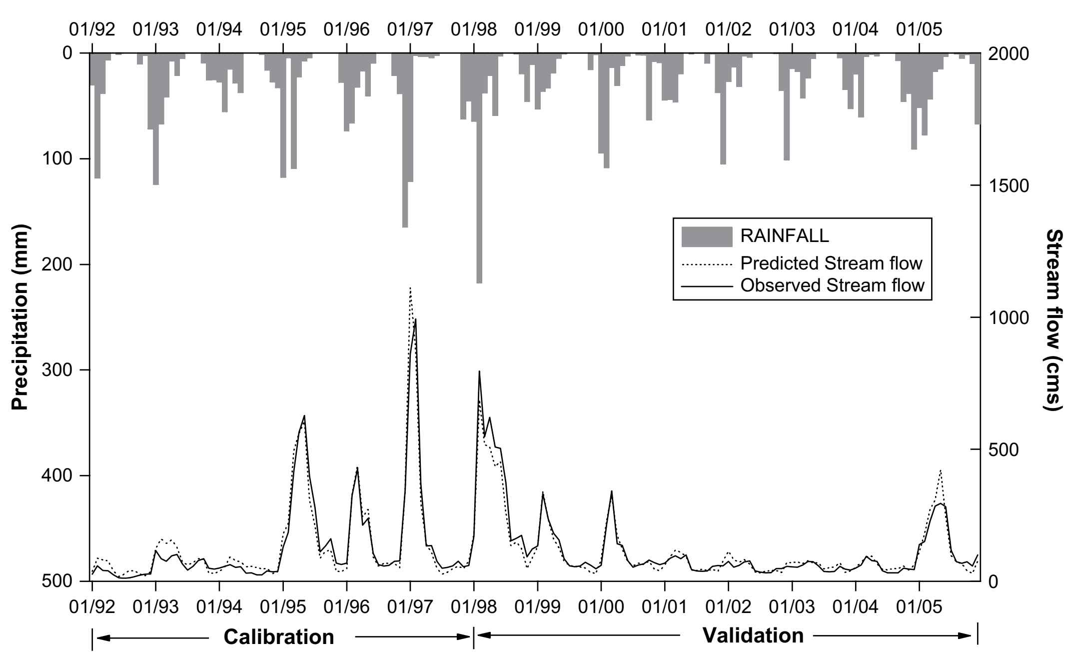

Fig. 2. Observed and predicted monthly stream flows for the San Joaquin River at Vernalis (USGS site #11303500) and monthly precipitation observed in Modesto, CA.

Table 5
Statistical results comparing observed and predicted monthly stream flow at selected sites during 1992–2005

Tributary or river sites	Calibration (1992–1997)			Validation (1998–2005)		
	NS	R^2	RMSE	NS	R^2	RMSE
Merced River[a]	0.83	0.87	10.2	0.67	0.78	8.8
San Joaquin River at Newman	0.91	0.94	40.6	0.88	0.90	38.6
Orestimba Creek	0.50	0.68	1.2	0.49	0.51	1.7
San Joaquin River at Cross Landing[b]	0.88	0.89	36.0	0.82	0.87	25.7
Del Puerto Creek	0.67	0.71	0.4	0.52	0.56	0.7
Tuolumne River	0.98	0.99	8.7	0.99	0.99	4.6
Stanislaus River	0.98	0.98	4.8	0.95	0.96	4.6
San Joaquin River at Vernalis	0.94	0.94	44.7	0.95	0.95	31.1

NS is the Nash–Sutcliffe coefficient, R^2 is the coefficient of determination, and RMSE is the root mean square error (m^3/s).

[a] For Merced River outlet (USGS #11273500), the period of record is 1992–1995 and 2002–2005.

[b] For San Joaquin River at Cross Landing (USGS #11274550), the period of record is 1996–2005.

decreased from 27.4 m^3/s at the boundary of the study area (USGS site #11270900), to 7.7 m^3/s near the confluence to the San Joaquin River (#11272500). Model results indicated that the SWAT model generates better hydrologic simulation for a subbasin, which received irrigation water from external sources, compared to that for a subbasin diverting irrigation water from the water bodies within the same subbasin.

SWAT model simulation also provided satisfactory stream flow prediction for the ephemeral streams on the western side of the watershed, which convey surface runoff from the Coast Range during winters and contain mostly agricultural surface drainage during the irrigation months. Deviations between observed and predicted stream flow were mainly found during irrigation seasons. In addition to precipitation and upstream inlet discharges, water diversions and agricultural drainages from irrigation return flow were the dominant factors in determining the temporal trends of stream flow rates in the San Joaquin Valley (Schoups et al., 2005). Due to the lack of data on actual irrigation water use for the study area and simulation period, the automatic irrigation operation in the SWAT was activated to estimate water amount delivered from streams or external sources and used in irrigation. The irrigation algorithm in the SWAT model limited the irrigated water amount by the soil field capacity, implying an assumption of high efficiency in water use and water diversion. This assumption might underestimate the agricultural drainage towards streams during intensive irrigation seasons. In general, the evaluation of model performance in Table 5 indicated that the SWAT model generated satisfactory simulation on stream flow in agricultural watersheds with minimal information on actual amounts of irrigation and water diversion.

Pesticides with high soil adsorption coefficients are more likely to get adsorbed to suspended sediments in stream flow. Therefore, reasonable estimation of sediment concentration in a stream was the first necessary step in simulating pesticide partitioning between dissolved and particulate phases. Fig. 3 shows the time-series plot of observed and predicted monthly sediment loads at the USGS site #11303500 (San Joaquin River at Vernalis). The trend of simulation followed the measured data, with Nash–Sutcliffe coefficients of 0.74 for both calibration and validation periods.

For other monitoring stations in the study area, sediment sampling records were only available for parts of the simulation periods. For a specific site, SWAT-predicted monthly sediment concentrations were compared with measured concentrations during the periods of record (Table 6). The predicted sediment concentrations fell within the ranges of the observations in terms of means and variances. It could be argued that the SWAT simulations were satisfactory in predicting the temporal trends and spatial variability of sediment yields given the rainfall and cropping pattern in the northern San Joaquin Valley watershed.

3.3. Modeling results of pesticides

3.3.1. Annual trends

Fig. 4 illustrates SWAT-predicted annual total dissolved loads of diazinon and chlorpyrifos at the watershed outlet (USGS site #11303500, San Joaquin River at Vernalis). Compared with the corresponding measured loads, the annual trends of pesticide loadings simulated by SWAT matched the measured data during

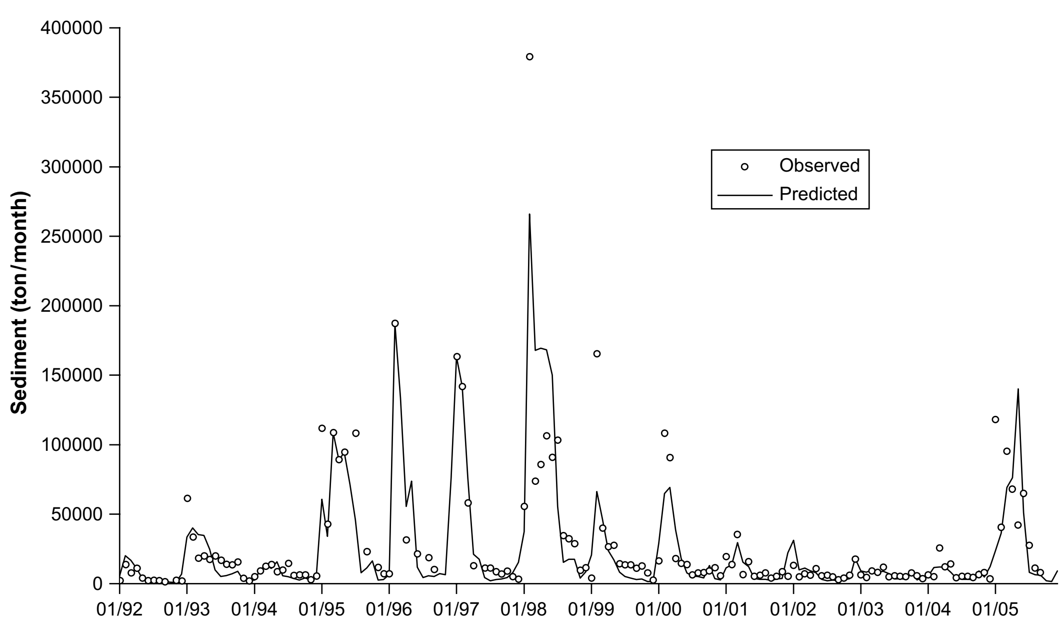

Fig. 3. Observed and predicted monthly sediment loads for the San Joaquin River at Vernalis (USGS site #11303500).

Table 6
Observed and predicted sediment concentrations (mg/L) in the northern San Joaquin Valley watershed

Tributary or river sites	Period of record	Observation		Prediction	
		Mean	Standard deviation	Mean	Standard deviation
Merced River	02/1997–08/2005	17.7	12.6	16.7	11.2
Orestimba Creek	02/1997–08/2005	167.5	147.6	154.7	164.4
San Joaquin River at Cross Landing	07/2000–11/2001	72.6	16.8	77.3	22.0
Tuolumne River	01/1993–03/1995	15.2	9.5	15.7	13.1
Stanislaus River	01/1993–09/1994	15.7	10.2	20.1	20.3

validation period of 1998–2005 (NS = 0.84 for diazinon and 0.77 for chlorpyrifos, respectively). Further analysis indicated that model performance in predicting pesticide loads were related to that in predicting stream flows over the simulation years. For example, predicted diazinon and chlorpyrifos loads in 1997 were significantly lower than measured data. This might be associated to the errors in predicted stream flow, especially during rainfall months as shown in Fig. 2. The modeling result confirmed that transport

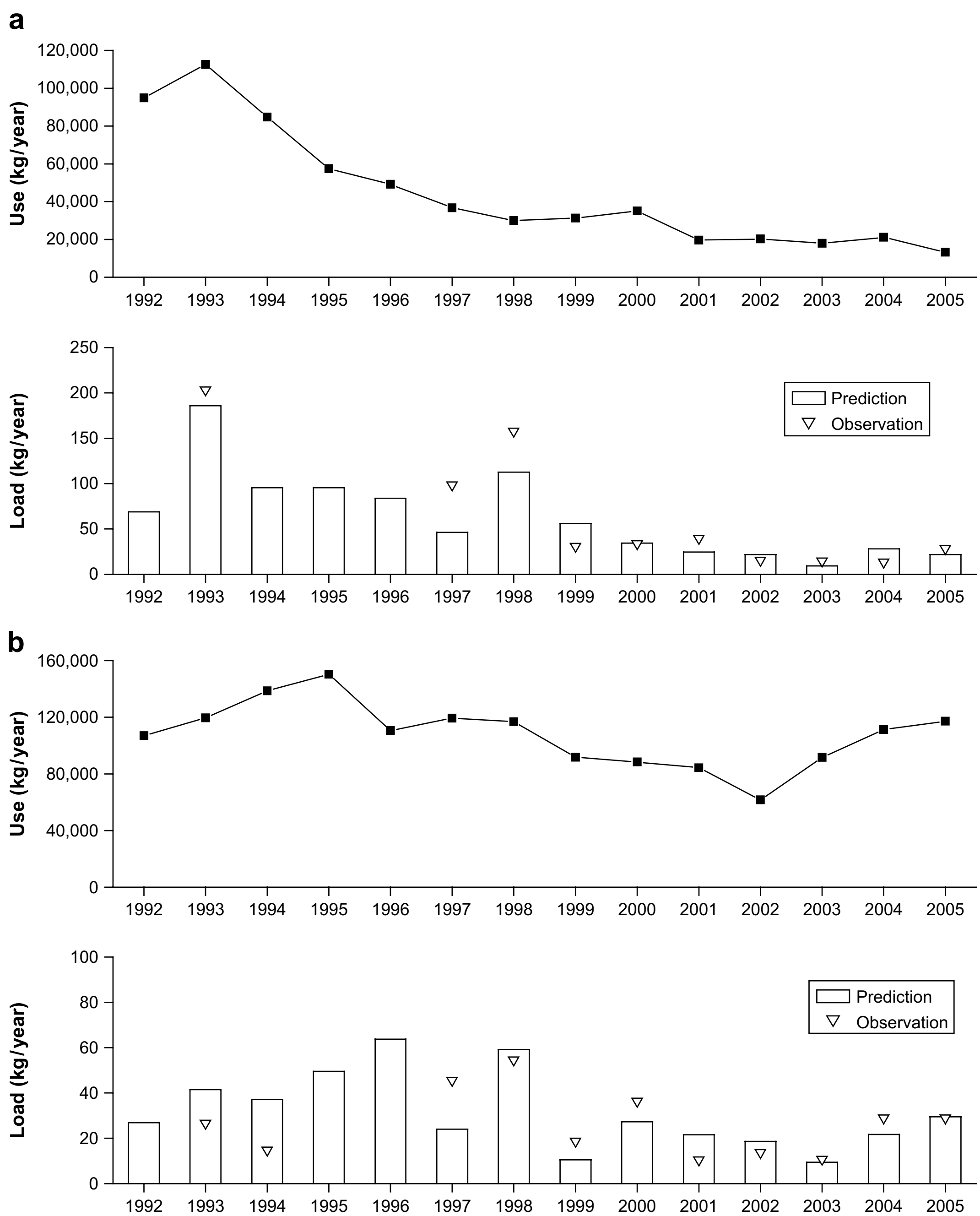

Fig. 4. Annual pesticide uses in the northern San Joaquin Valley watershed, and annual dissolved pesticide loads for the San Joaquin River at Vernalis (USGS site #11303500), for (a) diazinon and (b) chlorpyrifos during 1992–2005.

processes in surface runoff and stream flow were the governing processes in the pesticide fate and distribution. Comparisons of predicted and observed annual loads were also conducted at the outlets of Merced River and Orestimba Creek where records of pesticide measurements were available during the validation period of 1998–2005. The simulation of annual dissolved loads of diazinon and chlorpyrifos presented good or satisfactory agreement with measured data, as shown in Table 7. It is worth noting that the observed annual loads were estimated from incomplete records of field measurements taken at irregular intervals. In addition, for samples with concentration lower than the detection limit (0.005 μg/L for diazinon and chlorpyrifos), the concentration was recorded by the agency as the limit or an estimated value. Therefore, those observations used in the comparison should only be considered as reference values of the actual dissolved pesticide levels in the streams.

At the outlet of the studied watershed, the SWAT model predicted an average of annual in-stream diazinon loads of 96.0 kg during 1992–1997 and 38.5 kg during 1998–2005, respectively. The predicted load accounted for approximately 0.16% of the total agricultural applications of diazinon in the northern San Joaquin Valley watershed over the simulation period. This finding was in agreement with those reported by Domagalski and Munday (2003) and Kratzer et al. (2002), in which the ratio of the accumulated in-stream load and the total use of diazinon was 0.17% for both January–February 2000 and April–August 2001 in the San Joaquin River at Vernalis. For chlorpyrifos, the average annual loads at the watershed outlet were 40.4 kg and 24.7 kg, respectively, during 1992–1997 and 1998–2005, accounting for 0.03% of total application. The differences of pesticide loads between the two periods were primarily related to the pesticide use rates. For the periods of 1992–1997 and 1998–2005, averages of annual use rates were 72.6 and 23.5 tons for diazinon, and 124.2 and 95.4 tons for chlorpyrifos, respectively. The correlation between annual pesticide use and predicted load was strong for diazinon ($p = 0.001$) and moderate for chlorpyrifos ($p = 0.031$) during the simulation period (Fig. 4). In addition, average annual precipitation and stream flow, which were observed with higher values during 1992–1997 relative to those during 1998–2005, might contribute to the relatively smaller in-stream pesticide load during validation period.

Table 7
Observed and predicted annual loads of dissolved diazinon and chlorpyrifos at selected sites during 1998–2005

Statistical results for model evaluation[a]						
Tributary or river sites[b]	Diazinon			Chlorpyrifos		
	NS	R^2	RMSE	NS	R^2	RMSE
Merced River	0.69	0.85	1.31	0.55	0.79	1.53
Orestimba Creek	0.36	0.82	0.59	0.87	0.89	0.47
San Joaquin River at Vernalis	0.80	0.86	20.10	0.77	0.90	6.83

Means of annual dissolved loads (kg/year)				
Tributary or river sites	Diazinon		Chlorpyrifos	
	Observation	Prediction	Observation	Prediction
Salt Slough	7.76	6.62	5.45	6.62
Merced River	4.20	3.77	4.13	3.15
Orestimba Creek	0.79	0.84	0.50	0.44
Del Puerto Creek	0.52	0.44	0.31	0.10
Tuolumne River	5.90	4.71	3.36	3.14
Stanislaus River	4.39	5.71	4.26	4.43
San Joaquin River at Patterson	18.85	18.89	3.89	3.74
San Joaquin River at Vernalis	41.9	38.43	25.36	24.7

[a] NS is the Nash–Sutcliffe coefficient, R^2 is the coefficient of determination, and RMSE is the root mean square error (kg/year).
[b] Statistical results were not calculated for some tributary or river sites due to the data limitation.

3.3.2. Seasonal variation

Fig. 5 shows the monthly dissolved loads of diazinon and chlorpyrifos predicted by SWAT at the watershed outlet in comparison with the monitoring data. The Nash–Sutcliffe coefficients of the model performance on monthly pesticide simulation during 1992–2005 were 0.66 and 0.41 for diazinon and chlorpyrifos, respectively. The uncertainty in monitoring data might have significant effects on the evaluation of model prediction, especially at smaller time intervals. For example, the relatively low value of Nash–Sutcliffe coefficients for chlorpyrifos might be related to the low detection frequency of 42.1% (compared to 69.3% for diazinon) during the simulation period. Uncertainty might be introduced by replacing the measurements below detection limit with the limit or an estimated value, and be propagated in estimating the monthly loads and lower the statistical measures in model evaluation.

The SWAT model prediction indicated that transport of diazinon and chlorpyrifos in the study area was related to the timing of chemical applications and surface runoff. The predicted dissolved loads of diazinon at the watershed outlet were significantly correlated to the monthly uses over the watershed ($p < 0.001$). For the simulation period of 1998–2005, 51.5% of the diazinon application in the northern San Joaquin Valley watershed occurred in January and February, and 67.6% of overall amount of dissolved diazinon was predicted at the watershed outlet during those months with sizeable surface runoff. This finding was in agreement with the results of diazinon sampling during 1992–1995, which reported 74.0% of diazinon transport observed during January and February (Dubrovsky et al., 1998). For the growing and irrigation season of April to August, 35.5% of diazinon was applied within the watershed, whereas only 10.8% of dissolved diazinon load was predicted in this study at the watershed outlet.

Chlorpyrifos transport predicted by SWAT confirmed that the main factors involved in pesticide transport were timing of application and the occurrence of surface runoff. During the wet months of January and February, only 6.6% of annual chlorpyrifos application generated 28.5% of dissolved chlorpyrifos loads at the watershed outlet. Therefore, modeling results indicated that pesticide in-stream loads could be significantly reduced by decreasing pesticide use amounts during the wet months. During those months, the ratio between chlorpyrifos load and application was 0.12% for 1992–2005, which was comparable to the value of 0.16% reported by Kratzer et al. (2002) for the year of 2000. The correlation between reported uses and loads of chlorpyrifos was not significant. This might also be explained by the application timing of chlorpyrifos. The majority (76.8%) of chlorpyrifos application occurred during April–August. For those months, insufficient surface runoff and storms lowered the capacity and efficiency of pesticide yields towards surface waters, resulting in only 47.2% of annual amount of dissolved chlorpyrifos predicted at the watershed outlet.

3.3.3. Spatial distribution

SWAT model performance in predicting spatial distribution of dissolved diazinon and chlorpyrifos loads in the northern San Joaquin Valley watershed was evaluated in Table 7. The predictions were consistent with the observation ranges at tributary outlets and mainstem sites. It should be noted again that the observed annual loads estimated from measured pesticide concentrations were only presented as reference values due to the lack of continuous sampling data. For chlorpyrifos, the dissolved loads from subbasins were moderately correlated to the corresponding reported uses ($p = 0.008$) while the correlation was not statistically significant for diazinon ($p = 0.066$).

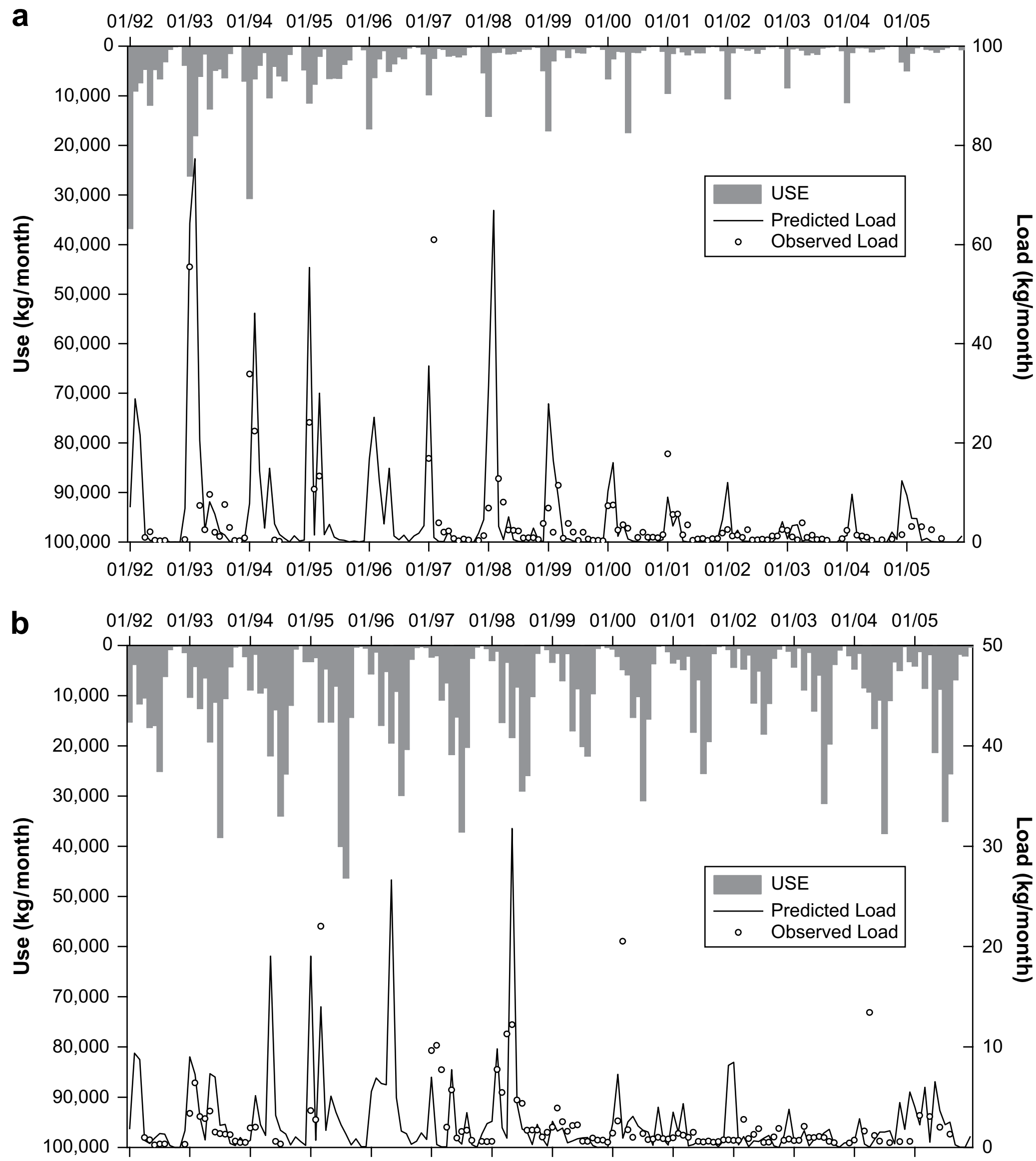

Fig. 5. Monthly pesticide uses in the northern San Joaquin Valley watershed, and monthly dissolved pesticide loads for the San Joaquin River at Vernalis (USGS site #11303500), for (a) diazinon and (b) chlorpyrifos during 1992–2005.

Fig. 6 shows the average pesticide yields, i.e., the ratio of pesticide load to the corresponding drainage area in the subbasins of the study area during 1998–2005. High pesticide yields were generally observed for the western subbasins, reflecting a reasonable spatial distribution pattern of pesticide yield towards the surface waters according to the conditions of hydrology, topography, and soil properties observed within the study area. Based on the soil and elevation data used in this study, subbasins on the western side of the San Joaquin River usually contain heavier textured soils with higher clay content and steeper slopes compared to the eastern side of the valley. Therefore, the western subbasins of the San Joaquin River produce more surface runoff than those on the eastern side. According to the simulation results, for example, the agricultural area in the subbasin of the Hospital Creek and Ingram Creek produced about 40% more surface runoff per unit area of that in the Stanislaus River subbasin with similar climate conditions during 1998–2005. The Salt Slough, one of the western tributaries, was identified as the tributary with maximum annual loads of dissolved pesticides discharged towards the San Joaquin River during 1998–2005, accounting for 17.2% and 26.8% of the loads at the watershed outlet at Vernalis, for diazinon and chlorpyrifos, respectively. Those areas with high pesticide yields could be candidates for further management evaluation to minimize the pesticide contamination in surface water. Preventative and mitigative management practices for pesticide should also be focused on those areas.

Stream flows in the western tributaries are dominated by agricultural drainage for much of the year and drains a western low-lying part of the study area with large areas of wetlands and agricultural land. Due to their low stream flow rates, therefore, the western streams showed high concentration of dissolved diazinon, which may have adverse effects on the organisms in the aquatic ecosystem. For example, at the Orestimba Creek outlet the simulation results showed that 55% of diazinon concentrations on a daily basis were above the detection limit of 0.005 μg/L, and about 3% of them exceeded 0.100 μg/L, a (4-day average) concentration shown to be toxic to aquatic life (CEPA, 2006). For the eastern tributaries of Merced, Tuolumne and Stanislaus Rivers, the predicted pesticide

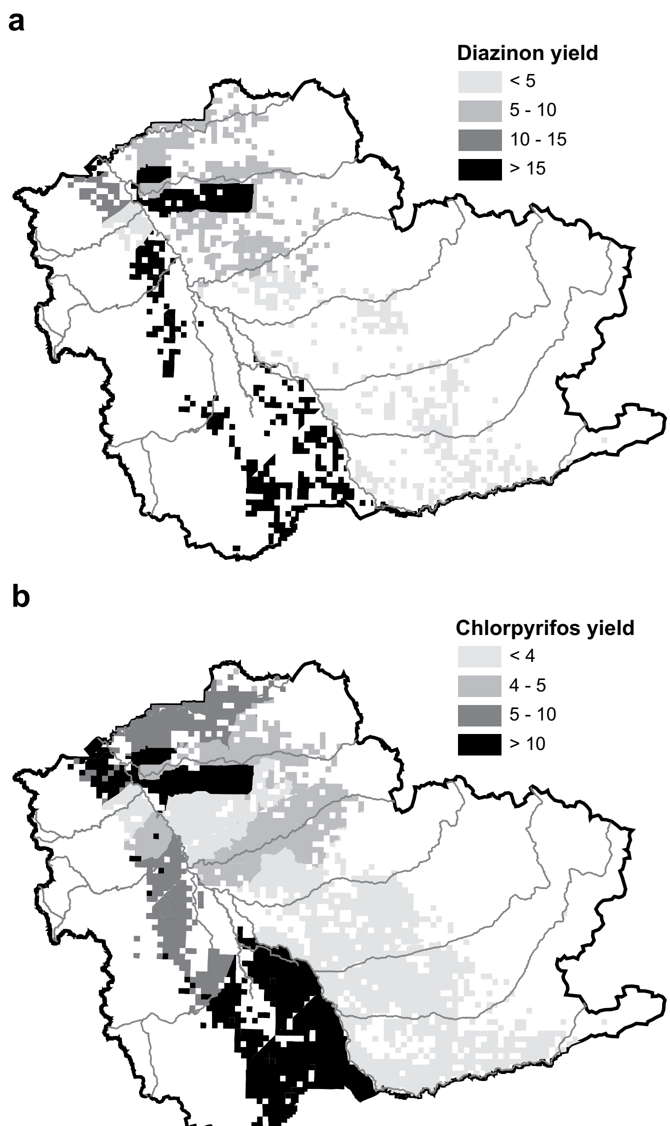

Fig. 6. Average pesticide yields (g/km^2) in the subbasins of the northern San Joaquin River watershed during 1998–2005 for (a) diazinon and (b) chlorpyrifos.

concentrations were much lower than corresponding levels in western side tributaries. The three eastern side tributaries carry runoff from the Sierra Nevada mountains year-round and pesticides are diluted by the upstream flows that were assumed to be pesticide-free in this study. Those water sources play important roles in improving the water quality in the San Joaquin River and its tributaries especially during rainfall seasons.

4. Summary and conclusions

The SWAT model was evaluated in the field conditions of the northern San Joaquin Valley watershed, California. The simulation of stream flow and sediment were first calibrated for the period of 1992–1997 and then validated for 1998–2005. Comparisons of predicted and observed data indicated that the calibrated SWAT model could be reliably used to simulate monthly stream flow and sediment transport in the subbasins of San Joaquin River and its tributaries.

Simulation components of surface hydrology and sediment transport in the SWAT model facilitate an efficient simulation of pesticide transport and fate at the watershed scale. This study presented comprehensive analyses of the temporal trend and spatial distribution of dissolved diazinon and chlorpyrifos loads in the San Joaquin River and its tributaries. The model performance was evaluated in terms of annual trend, seasonal variation, and spatial distribution in the dissolved loads of diazinon and chlorpyrifos at multiple sites within the study area. Statistical results indicated that the model generated good or satisfactory predictions compared to the monitoring data. The importance of surface hydrology, sediment transport, and physiochemical properties on predicting spatial and temporal variabilities on pesticide load in surface water was investigated by sensitivity analysis. On the basis of the model simulation and associated sensitivity analysis, three sets of factors primarily influencing the magnitude and trend of organophosphate pesticides loads in agricultural watersheds were identified: (1) magnitude and timing of surface runoff or agricultural drainage, (2) magnitude and timing of pesticide applications, and (3) physiochemical properties related to pesticide fate in soils. Modeling results of pesticide transport indicated that effective water quality control of pesticide contamination in the study area could be achieved by agricultural management practices.

In conclusion, the SWAT model exhibited the capability to simulate complex agricultural watersheds with appropriate calibration. The calibrated model in this study provided satisfactory simulation results in estimating temporal trend and spatial variation of stream flow, sediment, and dissolved diazinon and chlorpyrifos loads on monthly and annual bases in the northern San Joaquin Valley watershed. Therefore, the model is suitable for evaluating agricultural management practices and the associated environmental effects on water quality. The results of this study were instructive for further use of SWAT as an assessment tool in evaluating long-term trends and spatial patterns of pesticide transport in large agricultural watersheds in California's Central Valley and other regions of the United States.

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