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RESEARCH PAPER

Miki Sudo · Takuya Okubo · Ryoichi Kaneki

Paddy herbicide inputs in the entire river inflow reaching Lake Biwa, Japan

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Abstract This study estimated the inputs of four paddy herbicides in the entire river inflow reaching Lake Biwa, the largest lake in Japan, which serves as a water resource for 14 million people. The Uso River and the Hino River, the main contaminated rivers among the inflow rivers, were selected as daily and hourly monitoring sites to provide data on the seasonal trends in the concentration and load of herbicides and to determine the effect of rainfall events on load. The monitoring was also performed four times in 15 inflow rivers. The total input to the lake was calculated from the loads during fine weather conditions and additional loads during rainfall events. The former based on the lumped load from the two rivers and by prorating for the 15 rivers, and the latter was estimated from the relation between precipitation and increased load rate. The annual losses of herbicide from the basin to Lake Biwa were estimated to be 14.5% for bromobutide, 3.0% for pretilachlor, 5.2% for molinate, and 8.8% for simetryn. The loads caused by rainfall events accounted for 9%–18% of the total annual loads.

Key words Paddy herbicide · Water contamination · Rainfall event · Lake Biwa

Introduction

The use of pesticides contributes to high cultivation yields, high quality of agricultural products, and effective farm management. Because pesticides are generally synthetic chemical substances with toxicity, they can diminish the quality of drinking water and endanger nontarget species. Therefore, pesticide contamination of freshwater is a concern with respect to its long-term and low-dose effects on public health and the aquatic ecosystem. Rice is a major crop in Japan and is cultivated in an area totaling more than 17 000 km^2 each year (JSA 2002). Herbicide, fungicide, and insecticide are applied to paddy fields, and these substances are transported to rivers by surface runoff or infiltration. Paddy fields are one of the most important nonpoint sources of water pollution by pesticides. In recent years, many researchers have measured the herbicide contamination from paddy fields (Tanabe et al. 2001; Crepeau and Kuivila 2000; Albanis et al. 1994; Pereira and Hostettler 1993; Maru 1985).

Lake Biwa is the largest lake in Japan. In the catchment area, paddy fields occupy 17% of the total land. The water from Lake Biwa and the Yodo River, which receives water from the lake, is used by 14 million people as a source of potable water and for industrial, recreational, and agricultural purposes. With regard to the outflow from the lake, the concentration (Fukushima et al. 1995) and the amount of pesticide (Sudo et al. 2002a; Fukushima et al. 1995) released from Lake Biwa to the basin downstream were studied at the Seta River, which is the only natural outlet and flows from the southern end of the lake. Accurate evaluation of pesticide inputs from inflow rivers is required to understand the transport and fate of pesticides in the Lake Biwa basin. Tsuda et al. (1997) found that the maximum concentration in seven inflow rivers was 7.6 $\mu g\,l^{-1}$ for bromobutide, 8.7 $\mu g\,l^{-1}$ for pretilachlor, 75.5 $\mu g\,l^{-1}$ for molinate, and 21.2 $\mu g\,l^{-1}$ for simetryn. Sasagawa et al. (1996) reported that the maximum concentration in two small inflow rivers was about 10 $\mu g\,l^{-1}$, 400 $\mu g\,l^{-1}$, and 50 $\mu g\,l^{-1}$ for pretilachlor, molinate, and simetryn, respectively. Sudo et al. (2002b) estimated that the annual losses of pesticide to Lake Biwa in 1997 under fine weather conditions were 1.7% for esprocarb, 4.8% for mefenacet, 2.3% for thiobencarb, 3.7% for molinate, 9.4% for simetryn, and 13.0% for isoprothiolane. Several researchers have shown that rainfall events are a major source or, in some cases, the only source of pesticide pollution from upland fields, orchards, and golf courses (Dabrowski et al. 2002; Takahashi et al. 2000; Cryer

M. Sudo (✉) · R. Kaneki
School of Environmental Science, University of Shiga Prefecture, 2500 Hassaka, Hikone 522-8533, Japan
Tel. +81-749-28-8334; Fax +81-749-28-8477
e-mail: sudo@ses.usp.ac.jp

T. Okubo
Lake Biwa Environmental Research Institute, Otsu, Japan

et al. 1998; Sudo and Kunimatsu 1992; Wauchope 1978). However, none of the previous studies on the Lake Biwa basin have addressed the contribution of rainfall events to paddy pesticide loads.

The objective of this study is to elucidate the annual fluxes of four widely used paddy herbicides in the Lake Biwa basin with a detailed account of loads during rainfall events. To address this problem, field studies were performed in the Uso River and the Hino River, which are the major contaminated rivers and have large paddy field areas in their catchments, to investigate seasonal changes in the concentration and load and to study the effects of rainfall events on herbicide load. Basin-wide surveys were also performed to calculate the contribution of these two rivers to the loads of the entire inflow river system.

Materials and methods

Study area

Lake Biwa is the largest lake in Japan and is located in central Honshu, about 300 km to the west of Tokyo. Although more than 100 rivers originating in the surrounding mountains flow into the lake, the only natural outlet is the Seta River, flows which from the southern end of the lake with an average flow rate of $90\,m^3 s^{-1}$. Water also flows out through three artificial canals at an average flow rate of 20–$50\,m^3 s^{-1}$. The lake has a surface area of $674\,km^2$ and a basin that measures $3174\,km^2$, which closely corresponds to the administrative limits of Shiga Prefecture. The climatic conditions of the Lake Biwa basin are characterized by an average annual precipitation of 1740 mm and an average temperature of 14.1°C (LBRI 1989).

With more than $400\,km^2$ of paddy fields, rice is a major crop in the Lake Biwa basin and is harvested annually. The predominant geology in the paddy fields of the basin is unconsolidated sediments and the main soil types are gray lowland soils or gray soils. Rice seedlings are transplanted between the end of April and the beginning of May and harvesting is complete by mid-September in the majority of the paddy fields. Paddy fields are flooded in May and June, followed by intermittent irrigation throughout the growing period, except for mid-summer drainage in late June or early July. In most of the basins, individual paddy fields are irrigated with water from a river and a dam site by irrigation canals or pipeline supplies. Repeat irrigation is performed by supplying water collected from drainage, and an intake of the lake water is also supplied as irrigation water. Preemergence herbicides are applied by sequential treatment or by a one-shot treatment up to 3 weeks after transplantation. In sequential treatment, a first-stage herbicide is applied up to 5 days after transplanting, followed by a second-stage herbicide application between the second and third week. In the one-shot treatment, a one-shot herbicide is applied once between 3 and 14 days after transplanting. A second-stage herbicide is often applied additionally if the one-shot herbicide is not effective.

Water sample collection

The location of the observation sites is shown in Fig. 1. In this study, three types of observations were involved: (1) regular monitoring surveys during the harvest season, (2) intensive monitoring surveys during rainfall events and (3) basin-wide surveys in 15 inflow rivers.

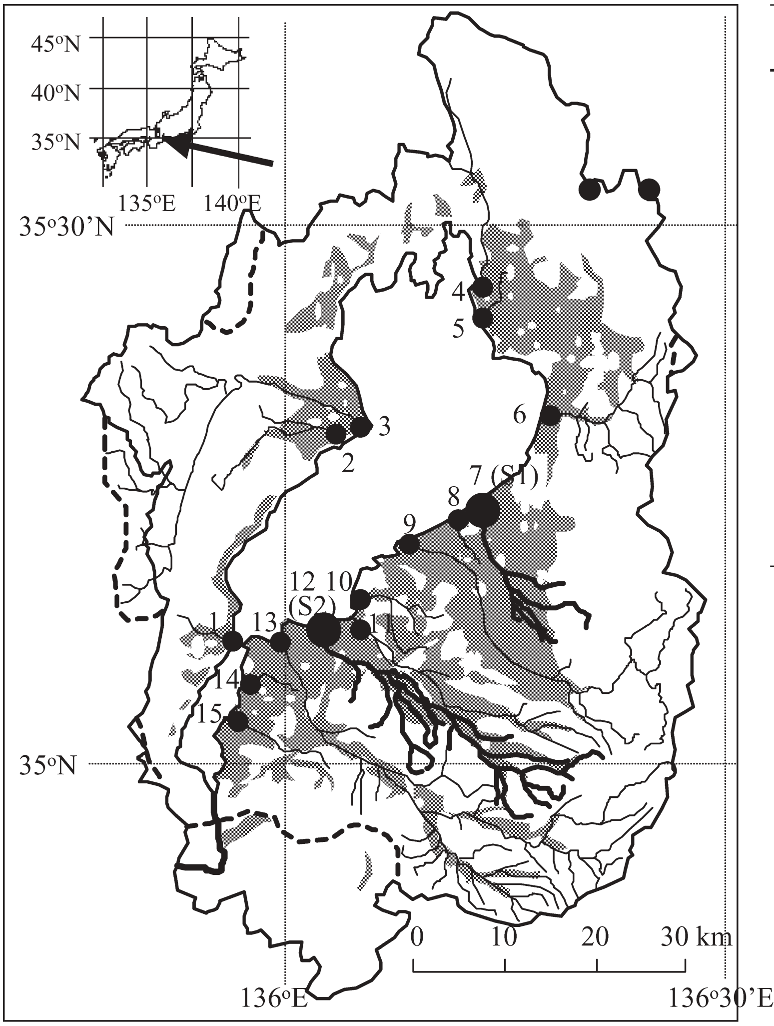
A previous study measuring pesticide concentration and load in 65 inflow rivers in 1997 showed that the significantly contaminated sites were the Uso River (S1) and the Hino River (S2); the sum of pesticide transport from the two rivers contributed 14% to 48% of the total mass from 65 rivers (Sudo et al. 2002b). The basins of the Hino and Uso include the third- and the fourth-ranking paddy field areas, respectively, of all the inflow rivers. Therefore, the two rivers were chosen as sites for a regular monitoring survey and an intensive monitoring survey. The two rivers run through paddy fields and small towns after flowing through a forested area, and then flow into Lake Biwa. Land utilizations are shown in Fig. 1. The water-stage heights at S1 and S2 were recorded at 10-min intervals. By calibration functions delineating the relation between stage height and discharge volume, after more than ten observations at various stage heights, each measured height was related to a discharge volume. The daily and hourly precipitation amounts were calculated from the average of precipitation collected at five locations within the watershed of the Hino River and six locations within the Uso River (Shiga Prefecture 2002).

In the regular monitoring surveys, water samples were collected over 8 months in 2001 (April–November), covering the herbicide application period and the rice harvest season. Sampling was conducted almost every day from April 15 to June 30, once or twice a week from July 1 to August 15, and once or twice a month for the remaining period. For the intensive monitoring surveys, water samples were collected before the onset of rains and during the following rainfall event at 2- to 12-h intervals. The hydrological conditions of monitored rainfall events are shown in Table 1. In both surveys, water samples were often collected with automatic samplers (ISCO 6700, Isco, Lincoln, NE, USA).

In the basin-wide survey, water samples were collected near the mouths of 15 influent rivers (Fig. 1.), selected on the basis of the areas of watershed and paddy fields, on May 26, June 16, and July 31 in 2000 and on May 10 in 2001. Water discharge at the time of sampling was also measured onsite. Because the precipitation monitored for 3 days before each observation was less than 2 mm, samplings were conducted at or near base flow conditions. Land utilization for the 15 rivers is shown in Fig. 1. The area of paddy fields in the basins of the 15 rivers corresponds to 54% of total paddy area in the Lake Biwa basin.

Sample analysis

The herbicides studied were bromobutide, pretilachlor, molinate, and simetryn. Their chemical names and physical



Site	River	Total area[a]	Land use[a] Paddy fields	Forests
		km^2	km^2	km^2
1	Mano R.	18.8	5.1	10.7
2	Kamo R.	43.4	6.1	34.8
3	Ado R.	309.7	11.3	282.9
4	Yogo R.	73.9	22.7	42.7
5	Chyonoki R.	11.6	9.6	0
6	Amano R.	111.0	21.6	74.2
7	Uso R.	84.2	43.8	22.3
8	Bunroku R.	8.1	5.8	1.0
9	Echi R.	203.3	19.1	161.9
10	Chyomeiji R.	129.2	66.8	27.6
11	Shiratori R.	22.0	12.9	3.9
12	Hino R.	210.1	57.8	106.0
13	Yasu R.	385.3	73.1	229.2
14	Horyu R.	4.1	3.1	0
15	Hayama R.	8.9	2.5	2.1

[a] Integrated Planning of Water Resource Quality, Department of Environmental Engineering, Kyoto University (2000)

● : Sampling site of regular and basin-wide surveys

● : Sampling site of basin-wide survey

——— : prefecture boundary

- - - - : drainage basin boundary

▒ : paddy fields

Fig. 1. Location of observation sites

Table 1. Hydrological conditions of intensive monitoring surveys during runoff events

Uso River (S1) Sampling period	Total precipitation (mm)	Maximum intensity (mm h^{-1})	Hino River (S2) Sampling period	Total precipitation (mm)	Maximum intensity (mm h^{-1})
2000			2000		
May 27–May 30	36.8	5.6	May 31–June 3	29.7	6.5
June 8–June 10	16.8	6.8	June 8–June 11	22.2	5.0
June 12–June 14	13.8	2.8	June 12–June 15	15.0	3.0
June 16–June 19	29.0	5.8	June 16–June 19	42.0	6.2
June 22–June 25	59.4	5.4			
2001			2001		
April 28–May 1	5.4	1.2	April 29–May 2	7.7	1.8
May 1–May 4	20.8	1.8	May 2–May 4	23.7	7.0
May 22–May 26	75.8	9.5	May 30–June 2	17.2	3.3
May 29–June 1	19.5	2.7	June 5–June 8	28.8	7.7
June 4–June 7	29.5	5.5			

Table 2. Chemical name and physical properties of observed herbicides

Common name	Chemical name	Water solubility [$mg l^{-1}$ (°C)]	Vapor pressure [mPa (°C)]	$\log K_{ow}$[a]	S-TD_{50}[b] (days)
Bromobutide	2-Bromo-3,3-dimethyl-*N*-(1-methyl-1-phenylethyl)butylamide	3.54 (25)	74 (25)	3.62	31–64
Pretilachlor	2-Chloro-2′,6′-diethyl-*N*-(2-propoxy-ethyl)acetanilide	50 (20)	0.133 (20)	4.08	20–50
Molinate	*S*-Ethyl azepane-1-carbothioate	880 (20)	746 (25)	2.88	8–25
Simetryn	N^2,N^4-Diethyl-6-methylthio-1,3,5-triazine-2,4-diamine	450 (room temp)	0.09 (20)	2.60	52–179

Physical data from Kanazawa 1996
[a] Partition coefficient between octanol and water
[b] Half-life in soil

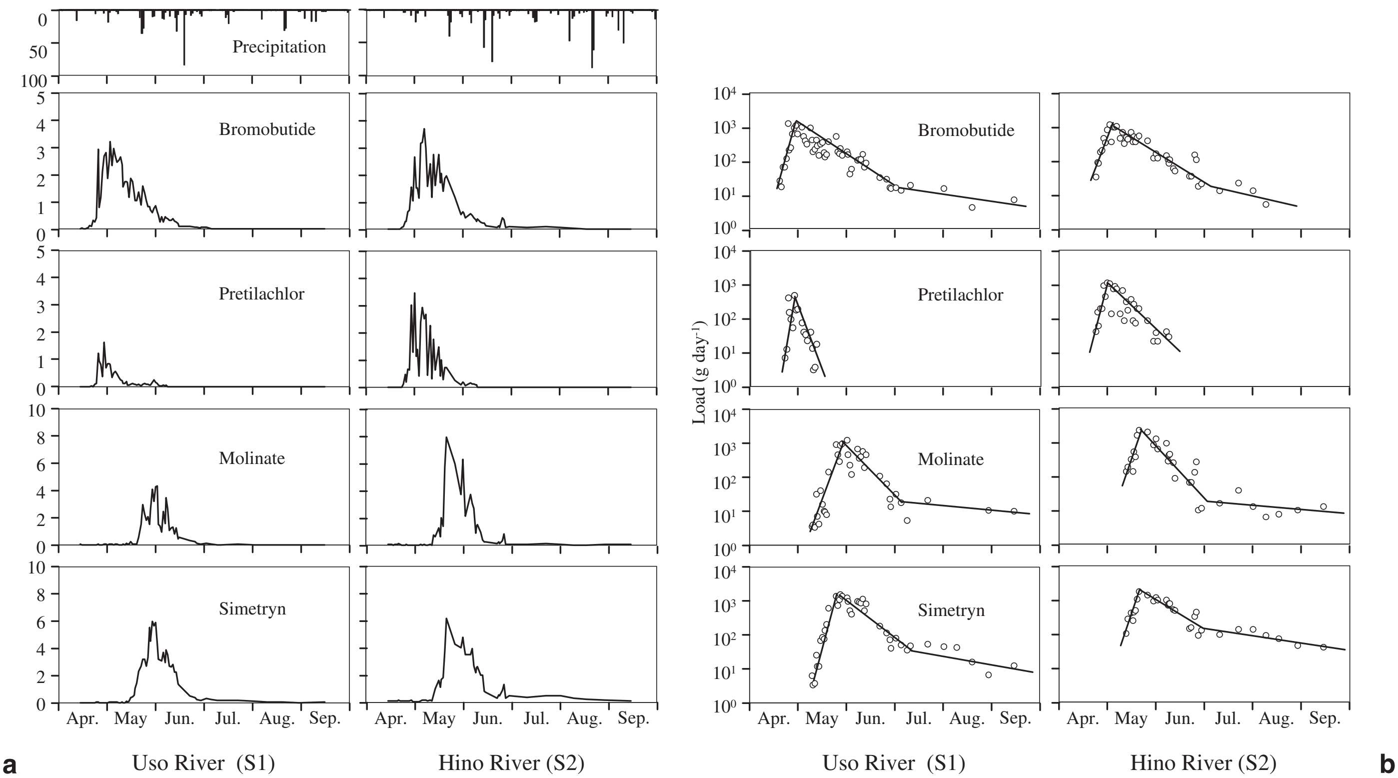


Fig. 2. **a** Herbicide concentrations (all data) and **b** loads under fine weather conditions at the Uso River (S1) and the Hino River (S2) during regular monitoring surveys (2001)

properties are listed in Table 2. Herbicide analytical standards and internal standards were obtained from Wako Pure Chemical Industries (Osaka, Japan) and Hayashi Pure Chemical Industries (Osaka, Japan). All chemicals used were of pesticide analytical grade. The herbicide concentrations were analyzed according to the method described in the previous study (Sudo et al. 2004). The limit of quantification for all compounds ($0.01\,\mu g l^{-1}$) was calculated by determining a concentration value that corresponded to an instrument signal/noise ratio of 5; the concentration of water samples detected at less than the limit was assigned a value of zero.

Results

Regular monitoring surveys in two inflow rivers

Herbicide concentrations in the water samples collected at the Uso River (S1) and the Hino River (S2) in the regular monitoring surveys are shown in Fig. 2a. Bromobutide and pretilachlor were applied as first-stage herbicides in sequential treatment between late April and early May. The concentrations of bromobutide and pretilachlor increased during their application period. The maximum concentra-

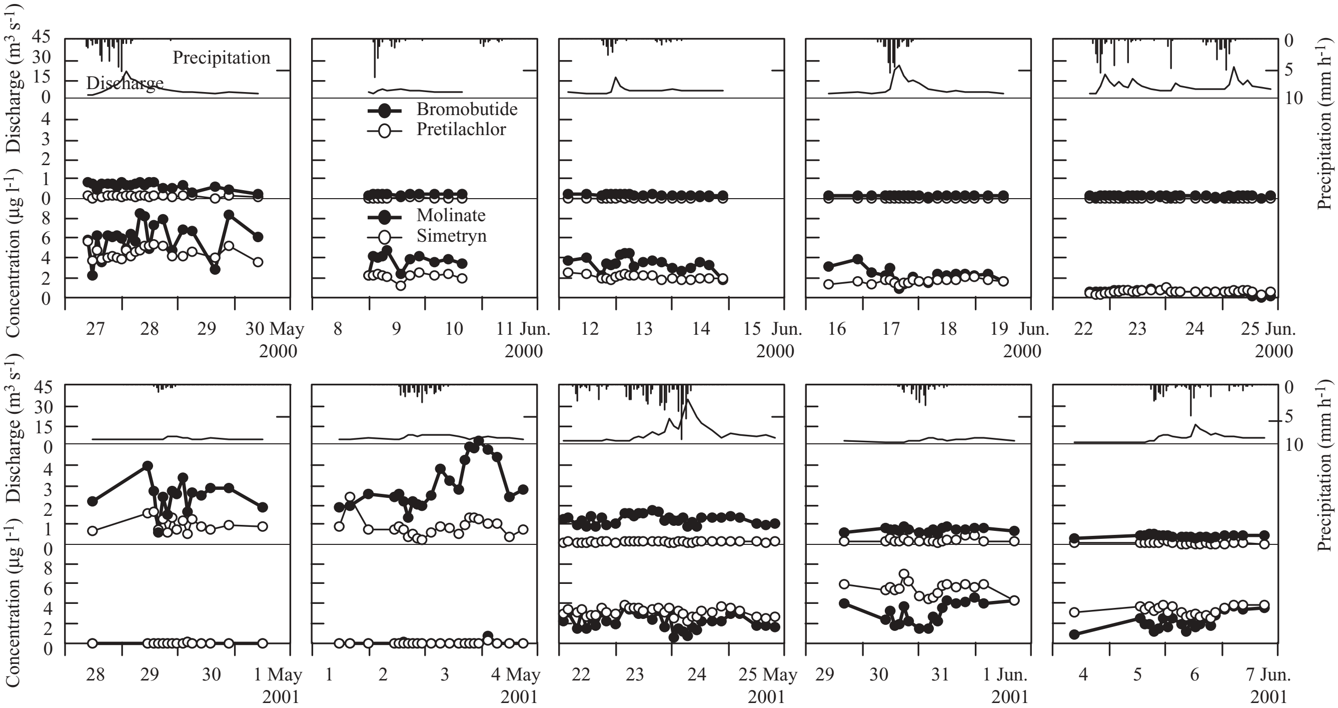


Fig. 3. Herbicide concentrations in the Uso River (S1) during rainfall events (2001). *Top*, discharge and precipitation; *middle*, concentration of bromobutide and pretilachlor; *bottom*, concentration of molinate and simetryn

tion of bromobutide at S1 and S2 was 3.2 and 3.7 $\mu g\,l^{-1}$, respectively. Subsequently the concentration gradually decreased and it was maintained at around 0.1 $\mu g\,l^{-1}$ in the summer season. The concentration of pretilachlor decreased rapidly after reaching a maximum of 1.6 and 3.4 $\mu g\,l^{-1}$ at S1 and S2, respectively. It was around 0.1 $\mu g\,l^{-1}$ in mid-June and could hardly be detected after July.

Molinate and simetryn were applied as second-stage herbicides in sequential treatment in late May. From mid-May onward, the concentrations of both molinate and simetryn increased rapidly. At S1 and S2, peak molinate concentrations of 4.3 and 7.9 $\mu g\,l^{-1}$ and peak simetryn concentrations of 6.0 and 6.2 $\mu g\,l^{-1}$, respectively were detected at the end of May. The concentrations then decreased to 1 $\mu g\,l^{-1}$ in mid-June and 0.2–0.5 $\mu g\,l^{-1}$ in July. The levels of both herbicides were at or near the detection limit during the remaining period.

Intensive monitoring surveys during rainfall events

As Fig. 2a shows, daily precipitation during the harvest season ranged from 1 to more than 80 mm. Herbicide concentration and load were observed in detail for ten events at S1 and eight events at S2. The daily precipitation in monitored events ranged from 5 to 76 mm at S1 and from 8 to 42 mm at S2 (Table 1). Figure 3 shows the herbicide concentrations at the Uso River during rainfall events. Also shown in Fig. 3, the water volume increased sharply in response to precipitation; however, herbicide concentrations were generally maintained at the pre-event level throughout the event, or they often decreased slightly at discharge peaks. The profile of herbicide concentrations in the Hino River was the same as that observed in the Uso River.

Basin-wide surveys in 15 inflow rivers

Table 3 shows the detection frequencies and the maximum herbicide concentrations in the samples collected from 15 inflow rivers. Higher contamination was found during the application period as observed in the regular surveys. Bromobutide and pretilachlor were detected in almost 90% of the samples in May; however, levels decreased to 67% and 53%, respectively, in late July. Molinate and simetryn were detected in almost all the samples throughout the year, except for those collected in early May, just before the application period.

Table 4 shows the ratio of the sum of the loads from the Hino and Uso rivers to that of the 15 rivers, according to the basin-wide surveys. The arithmetic mean values (R_L) show that the lumped load from the Hino River and the Uso River contribute 21%–44% to the total load from 15 rivers. This difference in the ratio of herbicide load may be mainly due to the application amounts in the basin.

Discussion

Seasonal trends in herbicide load under fine weather conditions were estimated from the data set of regular monitor-

Table 3. Detection frequencies and maximum concentrations in basin-wide surveys

Sampling	Discharge ($m^3 s^{-1}$)	Bromobutide		Pretilachlor		Molinate		Simetryn	
		Freq.[a] (%)	Max.[b] ($\mu g\, l^{-1}$)	Freq. (%)	Max. ($\mu g\, l^{-1}$)	Freq. (%)	Max. ($\mu g\, l^{-1}$)	Freq. (%)	Max. ($\mu g\, l^{-1}$)
2000									
May 26	0.07–3.72	93	2.10	87	1.60	100	37.2	100	18.1
June 16	0.16–5.77	93	0.31	73	0.21	100	2.85	100	3.54
July 31	0–5.55	67	0.08	53	0.13	93	0.21	87	0.60
2001									
May 10	0.08–4.42	87	6.77	93	5.13	40	0.27	40	0.30

[a] Frequency
[b] Maximum concentration

Table 4. Ratio of load from the Hino and Uso Rivers to that of the 15 rivers in the basin-wide surveys

Sampling	Discharge (%)	Bromobutide (%)	Pretilachlor (%)	Molinate (%)	Simetryn (%)
2000					
May 26	24	41	19	46	51
June 16	27	45	20	35	44
July 31	22	–[a]	–[a]	17	32
2001					
May 10	13	43	26	–[b]	–[b]
Average(R_L)[b]	–	43	21	33	44

[a] Excluded from the calculation of R_L values
[b] Arithmetic mean

ing surveys in the two rivers. The data collected under rainy conditions, identified by daily precipitation levels, were removed prior to performing the statistical analysis. The herbicide load was divided into three periods: the period before reaching peak load, during the peak load and the following high load, and during the low load. As shown in Fig. 2b, the load during each period was approximated by a straight line plotted against a semilogarithmic scale and expressed as:

$$L_B(t) = a_1 \cdot \exp(-kt) \quad (1)$$

where L_B is the herbicide load under fine weather conditions at time t, and a_1 and k are constants. April 1, 2001, marked the beginning of the sampling period ($t = 1$). The correlation coefficients (r^2) of the estimated regression lines were mostly greater than 0.8, although they were often less than 0.8 during the lower loading periods.

Herbicide applied to paddy fields remains in the soil surface layer and interacts with the incoming water. The leaching from paddy fields occurred vertically through the soil profile and horizontally under the levee. During fine weather, the main cause of herbicide transport is infiltration runoff, although a small part may be due to the surface runoff induced by artificial drainage, inadequate water management, and leakage from the levee. Thus herbicide remaining in paddy soil was discharged successively as shown in Fig. 2. During a rainfall event, herbicide outflow from the paddy fields may be accelerated to some extent by infiltration and runoff; however, a significant outflow from field outlets may occur only during heavy rainfall, when the water depth in paddy fields exceeds the height of the outlets. Even in such cases, rainwater overflows directly into the drainage system without interacting with the herbicide remaining in the soil. Thus the pesticide concentration in a drainage channel or river was affected in a minor way by rainfall events. In plowed fields, significant pesticide loss is induced temporarily by a rainfall event as indicated by previous studies (Dabrowski et al. 2002; Takahashi et al. 2000; Cryer et al. 1998; Sudo and Kunimatsu 1992; Wauchope 1978), either by dissolving in runoff water or being carried off as suspended soils in erosion.

Although the concentration of herbicide during rainfall events generally remained at the same level, an increase in the herbicide load was associated with the increase in water volume. The effects of a rainfall event on herbicide load are not easy to predict: multiple factors are involved such as the amount of rainfall, rainfall intensity, and proximity of the rainfall event to herbicide applications (Dabrowski et al. 2002; Besta et al. 1997; Scrunshaw and Lester 1995; Sudo and Kunimatsu 1992; Wauchope 1992). In this study, some generalizations were made. To eliminate the factor of the timing of the rainfall events, the net load during monitored events (L_{RN}) was calculated as follows:

$$L_{RN} = L_{RT} - L_B \quad (2)$$

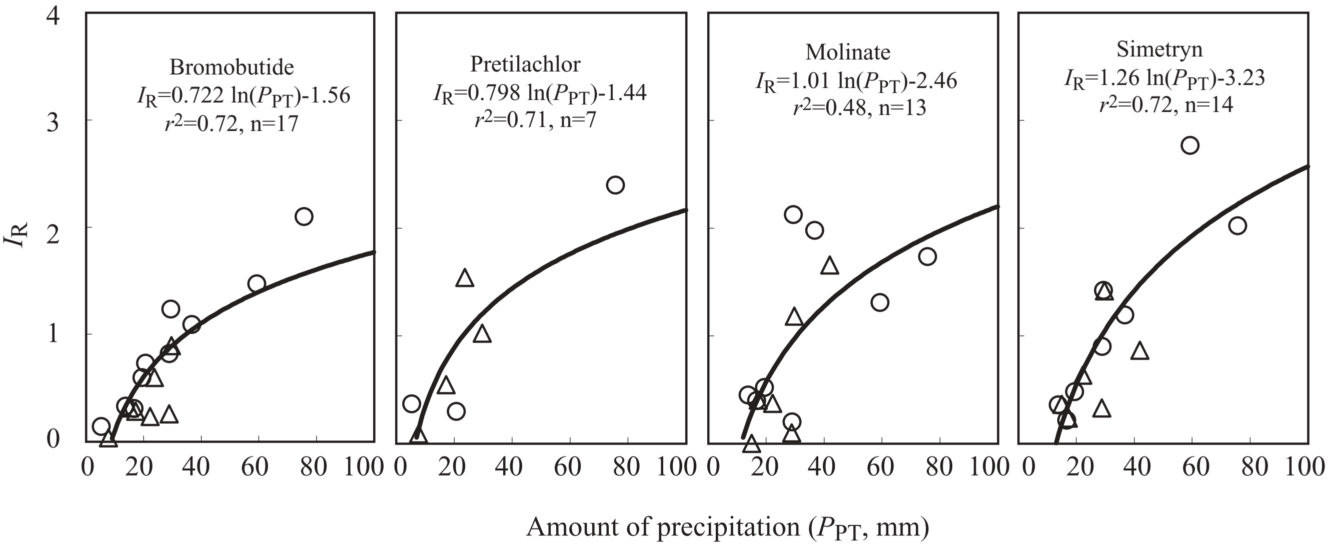


Fig. 4. Relationship between the increased rate of herbicide load during a rainfall event (I_R) and precipitation levels. *Circles*, Uso River; *triangles*, Hino River

where L_{RT} is the total herbicide load during an event and L_B is the expected load during fine weather, calculated using the load just before a rainfall event using Eq. 1. L_{RT} was calculated using Eq. 3, where C_i and Q_i were the concentration and the water volume monitored at sampling time t_i and n was the number of sampling.

$$L_{RT} = \sum_{i=1}^{n} C_i \cdot Q_i \left(t_{i+1} - t_{i-1}\right) \Big/ 2 \quad (3)$$

The increased rate of herbicide load during a rainfall event (I_R) was calculated as follows:

$$I_R = L_{RN} / L_B \quad (4)$$

Previous studies (Kunimatsu and Sudo 1997; Kunimatsu 1997) show that the relation between the precipitation amount and material load is a useful way to estimate the load during a rainfall event. Although this method cannot account for the effect of rainfall intensity on loads, it has the advantage of providing reasonable estimations without requiring a detailed and continuous water volume data set. Figure 4 shows the relationship between the value of I_R and the amount of precipitation. Data obtained during low- or no-contamination periods were removed prior to the analysis. Data from the Uso River and the Hino River were lumped together into a single correlation between I_R and the log-transformed precipitation amount (P_{PT}) as indicated in Eq. 5.

$$I_R = b_1 \cdot \log\left(P_{PT}\right) - b_2 \quad (5)$$

Figure 4 shows that the intersection with the longitudinal axis represents the practical precipitation affecting on the herbicide load. The I_R value increased in proportion to the precipitation, although the increase was attenuated by heavy rainfall for the reasons mentioned above. Figure 4 shows high correlation coefficients (r^2 > 0.7) for bromobutide, pretilachlor, and simetryn. The relatively low value of r^2 for molinate (0.48) may be caused by its high volatilization capacity based on a high Henry's constant (Armbrust 1999).

Annual herbicide inputs in 2001 from all the rivers flowing into Lake Biwa (L_{in}) were calculated from the data obtained from surveys at the Uso and Hino Rivers, as well as from the basin-wide survey as follows:

$$L_{in} = \sum \left(L_B(t) + L_R(t)\right) \quad (6)$$

where $L_B(t)$ is the daily mass load under fine weather conditions and $L_R(t)$ is the additional load caused by a rainfall event. $L_B(t)$ was calculated from the load for the Uso River (L_{Buso}) and the Hino River (L_{Bhino}) obtained using Eq. 1 and R_L as follows:

$$L_B(t) = \left(L_{Buso}(t) + L_{Bhino}(t)\right) \Big/ R_L \cdot A_T / A_{15} \backslash \quad (7)$$

where A_T is the total paddy area in the Lake Biwa basin, A_{15} is the total paddy area for the 15 rivers monitored in this study, and t represents the dates from April 1. By including the contribution of loads from two rivers through actual measurement, calculations using R_L can provide reasonable results compared to calculations based only on the proportional allocation of the area. Values for $L_R(t)$ were calculated using L_B and I_R obtained from Eq. 5 as follows:

$$L_R(t) = L_B(t) \cdot I_R\left(P_{PT}(t)\right) \quad \text{if} \quad I_R \le 0 \text{ then } I_R = 0 \quad (8)$$

where P_{PT} is the mean precipitation calculated from 13 points of AMEDAS data (JMA 2004) within the Lake Biwa basin. The estimation was continued until the herbicide concentration calculated from L_B was below the quantification limit.

Table 5 shows the annual losses expressed as a percentage of the herbicides applied to the basin. The amount of application was estimated to be 93% of the total shipment to the prefecture in 2001, corresponding to the ratio of the Lake Biwa basin to the prefecture. Although monitoring

Table 5. Annual loads and losses of herbicides from the Lake Biwa basin (2001)

	Application amount (tonne)	Load		Herbicide loss (% of application)
		Total (kg)	During rainfall (kg)	
Bromobutide	1.82	263	23	14.5
Pretilachlor	4.54	136	16	3.0
Molinate	5.13	269	44	5.2
Simetryn	4.52	396	70	8.8

frequencies and calculation methods are different from this study, previous field studies for Japan have shown that losses of pretilachlor, molinate, and simetryn were 2.7%–25.2% (HIES 2000; Inoue and Ebise 1999; Numabe et al. 1992), 2.3%–29.5% (Sudo et al. 2002b; Sasaki et al. 1994; Ishii 1984), and 7%–17% (Sudo et al. 2002b; HIES 2000; Nakamura et al. 1982), respectively. Conventional calculation methods for pesticide load, e.g., the simple integration of load obtained from weekly sampling, may lead to overestimation or underestimation because concentrations and loads vary on a daily basis.

Table 5 also shows the total herbicide inputs and inputs caused by rainfall events in Lake Biwa in 2001. As described previously, many studies show that a rainfall event is a vital source of pesticide input to rivers or lakes from plowed fields. However, in this study the loads of investigated herbicides caused by rainfall events accounted for only 9%–18% of the total loads. These results were induced by the flooded condition during the growing period, which is the primary difference between paddy fields and plowed fields.

Although physically based models have been used for prediction of paddy pesticide concentrations (Inao et al. 2001, 2003; Kibe et al. 2000; Mullins et al. 1993), they have been established only under small-scale and edge-of-field conditions. Very few empirical studies have shown a quantitative relation between watershed monitoring data and individual paddy pesticide properties. Such empirical relations would allow a better evaluation of the relative runoff for different compounds and provide quick estimates of potential contamination levels in watersheds. In this study, relations between the physical/chemical properties cited in Table 2 and pesticide losses both in fine weather and during rainfall events were not clearly elucidated. Possible reasons for this lack of correlation may be the heterogeneity of watershed factors, including soil properties, water management practices, and basin morphologic characteristics as well as the validity of pesticide application practices such as application duration and the formulation used.

Losses of herbicide obtained in this study may provide the basic data to estimate the effects on the usability of water and the aquatic ecosystem. The transport and fate of pesticides may also give a useful pointer to the movement of other chemicals in the environment. In any case, intensive long-term observation and the construction of predictive models are required because low contamination levels of pesticide may be a potential hazard to humans and the environment.

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