Original Article

Effect of Long-Term Successive Applications of Organic Fertilizers on Dissipation of Several Pesticides in Two Soils

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(Received July 28, 2003; Accepted November 10, 2003)

The dissipation rates of dimethoate, fenobucarb, flutolanil, simazine, prometryn and alachlor were compared among an andosol and a gray lowland soil subjected to different fertilizing practices over a 20 year period. The rate constants for dissipation of most pesticides per biomass carbon, biomass nitrogen and esterase activity among the plots in each soil were less variable than the corresponding rate constants, indicating that the dissipation depended on microbial amount and activity. The rate constants in the gray lowland soil were similar to or greater than those in the andosol, despite the smaller values of microbial amount and activity in the former. This is due to the larger water soluble fraction/acetone soluble fraction ratios in the former. The long-term succesive applications of organic fertilizers were less effective in the dissipation for the gray lowland soil than the andosol. This is likely to result from a less effective accumulation of microbial biomass in the former.

Keywords: long-term successive applications of organic fertilizers, andosol, gray lowland soil, dissipation rates of pesticides, microbial amount and activity.

INTRODUCTION

Public concern has been growing about the influence of agricultural practices on the environment. The use of organic materials is essential for the maintenance and improvement of soil fertility. Investigations have therefore concentrated mostly on changes in soil properties 1-3) and subsequent crop yields. 1-4) To be effective, pesticides need to persist for a certain period. However, the risk of environmental contamination becomes greater with an increase in the persistence of pesticides. The degradation rates of pesticides depend on microbial amount and activity. 5-7) The addition of organic fertilizers generally enhances these two biological properties, 8-10) and hence is expected to stimulate the degradation of pesticides. 9,11) Since microbial degradation is an important process controlling the fate of contaminants in soil systems, the addition of organic fertilizers should be evaluated from an environmental standpoint such as minimizing the exposure to pesticides of not only humans but other non-target organisms. The purpose of the present study is to clarify the effect of long-term successive applications of organic fertilizers on the dissipation of six pesticides using two soils varing in physical, chemical and biological properties, and to obtain information with which to carry out appropriate agricultural practices to minimize the risk of environmental contamination from pesticides.

MATERIALS AND METHODS

1. Chemicals

To make an overall assessment of the effect of organic fertilizers on the dissipation of pesticides, six pesticides, divided into five chemical families, were investigated; the insecticides dimethoate (*O*, *O*-dimethyl *S*-methylcarbamoylmethyl phosphorothioate) and fenobucarb (2-sec-butylphenyl *N*-methyl-carbamate), the fungicide flutolanil {*N*-[3-(1-methyethoxyphenyl]-2-(trifluoromethyl)benzamide}, and the herbicides simazine [2,4-bis(ethylamino)-6-chloro-1,3,5-triazine], prometryn [2,4-bis(isopropylamino)-6-methylthio-1,3,5-triazine] and alachlor [2-chloro-*N*-(2,6-diethylphenyl)-*N*-(methoxymethyl)acetamide]. The physicochemical properties of the pesticides are shown in Table 1. None of the six had been applied in the previous five years, and none were detected in any soil samples collected.

The pesticides and reagents used were purchased from Wako Pure Industries, Co., Ltd. (Tokyo). The purities of the chemicals were more than 97%.

2. Soil Samples

The soils were collected from two long-term experimental fields on amendment of organic fertilizers; the Tochigi Prefectural Agricultural Experiment Station at Utsunomiya,

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Table 1. Physicochemical properties for the six pesticides

Pesticide	es ^{a)}	Chemical family	Water solubility ^{b)} (mg/l)	t _{1/2} c) in soil (days)
Dimethoate	(I)	Organophosphorus	25,000	16-33
Fenobucarb	(I)	Carbamate	660	6-30
Flutolanil	(F)	Amide	9.6	208
Simazine	(H)	Triazine	6.2	72
Prometryn	(H)	Triazine	33	40-70
Alachlor	(H)	Anilide	242	42-70

^{a)} (I): insecticide, (F): fungicide, (H): herbicide. ^{b,c)} From Ref. 12.

and at Tochigi. The soils were classified as a highly fumic andosol at the Utsunomiya site and a gray lowland soil at the Tochigi site, each contributing to a large proportion of the arable land in Tochigi Prefecture. The fertilizer treatments were started in 1977 at both sites. Both fields have been used to grow barley (*Hordeum vulgare* L.) as a winter crop. In Tochigi, paddy rice (*Oryza sativa* L.) has been cultivated in the summer crop, while in Utsunomiya, soybean [*Glycine max* (L.) Merr.] and upland rice were grown every other year until 1997, and lettuce (*Lactuca sativa* L.) since 1998.

In early June 2000, following harvest of the barley, soil samples were collected from plots subjected to one of three different fertilizing practices; Utsunomiya, 1) the plot only received inorganic fertilizers every crop in the andosol (A-IF plot). Inorganic fertilizers were applied at recommended rates for the other plots as well, 2) the plot additionally received a compost consisting of cattle feces and rice straw at a recommended rate (1500 kg fresh weight/10 a for both crops, A-CM plot), and 3) the plot additionally received cattle feces at a rate of 2400 kg fresh weight/10 a for both crops, which is equivalent to the rate in the A-CM plot on a dry-weight basis (A-CF plot), and Tochigi, 1) the plot only received inorganic fertilizers every crop in the gray lowland soil (G-IF plot), 2) the plot additionally received compost consisting of cattle feces and saw dust at a recommended rate (1000 kg fresh weight/10 a for both crops, G-CM plot), and 3) the plot additionally received straw at normal rates (rice straw: 500 kg fresh weight/10 a for winter, and barley straw: 300 kg fresh weight/10 a for summer, G-SR plot).

For each plot, ten samples were collected from the plowing layer (15 cm depth) in the inter-row space. Half of each sample was carefully dried to 40% of maximal water holding capacity, ensuring that no part of the sample became airdry, then passed through a 2 mm sieve. Thereafter, the sample was moistened to 60% of maximal water holding capacity and pre-incubated for 10 days at 25°C to allow the metabolic activities to stabilize after the initial disturbance. The sample was used for analyses of pesticide residues, amount of microbial biomass and fluorescein diacetate (FDA) hydrolytic activity. The remainder was air-dried, passed through a 2 mm sieve, and used to analyze other soil

properties.

3. Analytical Procedure

3.1. Microbial biomass and activity

The pre-incubated soils, 10 g on a dry-weight basis, were used for the analyses of microbial biomass carbon and nitrogen. Values were determined by the chloroform fumigation-extraction method, followed by extraction with 0.5 M K₂SO₄. ¹⁴⁾ Microbial biomass carbon was evaluated by the dichromate oxidation technique, ¹⁴⁾ and microbial biomass nitrogen by the peroxydisulfate oxidation method ¹⁵⁾ followed by the hydrazine reduction technique. ¹⁶⁾

FDA is hydrolyzed by various enzymes, ¹⁷⁾ and the hydrolytic activity is positively correlated with microbial biomass ^{18,19)} and rate of respiration. ¹⁹⁾ The activity was measured according to the method of Schnürer and Rosswall1 ¹⁹⁾ for the evaluation of total microbial activity. The pre-incubated soils, 2 g on a dry-weight basis, were suspended in 100 ml of 60 mM sodium phosphate buffer (pH 7.6), and then 1 ml of a 1 mg/ml FDA acetone solution was added. The mixture was incubated at 24°C, and the absorbance at 490 nm was measured with a spectrophotometer (Hitachi Co., Ltd., U-3000) at 0, 1, 2 and 3 hr. The activity expressed as the increase in absorbance at 490 nm per unit of soil weight and time (ΔA_{490} /g • hr) was calculated using the least squares method.

3.2. Pesticide residues

The pre-incubated soils, 20 g on a dry-weight basis, were placed into 100 ml glass vials. The top of each vial was loosely wrapped with aluminum foil for air exchange. The pesticides were dissolved in an acetone solution to bring individual concentrations to 200 µg/ml. Two hundred microliters of the solution (equivalent to $2 \mu g/g$ on a dry soil basis) was added to the soils and mixed well using a glass rod. The soils were then incubated under the same conditions as for the pre-incubation. Losses from evaporation were compensated twice a week by adding distilled water. The residual amounts of pesticides in the soils were periodically analyzed for the acetone soluble fraction (ASF) and water soluble fraction (WSF) as reported previously.²⁰⁾ Analyses were made with duplicate samples at each time point; 1, 4, 7, 14, 28, 56 and 91 days after incubation. Prior to the analysis by gas chromatography (GC), a clean-up procedure for the ASF was carried out using a silica gel column for dimethoate and a florisil column for the other pesticides.

The pesticide residues were analyzed with a Hewlett Packard 6890 GC equipped with a fused silica capillary column (HP-5 MS, 30 m \times 0.25 mm i.d., 0.25 μ m film) / a Hewlett Packard 5973 MS operated in the selected ion monitoring mode. The detection limit was 0.005 μ g/g for dimethoate and simazine, and 0.001 μ g/g for the other pesticides.

The recovery test was carried out by the acetone extraction method; $2 \mu g$ of each pesticide was added to the soils. The recoveries of the six pesticides were 89–97% for the

c) $t_{1/2}$: half-life

G =

andosol and 94-106% for the gray lowland soil.

3.3. Data analysis

The dissipation rates of pesticides in the soils were calculated assuming the first order kinetics:

$$C = C_0 e^{-kt}$$

$$\ln C = \ln C_0 - kt$$

where C is the concentration of pesticide as a function of time t, C_0 is the concentration of pesticide at time 0, and kis the rate constant for dissipation in soil. The k values were calculated by fitting the observed data to the above equation using the least squares method. The half-lives $(t_{1/2})$ of pesticides were calculated from the equation $t_{1/2} = -0.693/k$.

RESULTS

1. Soil Properties

The soil textures were loam (clay 12.3%, silt 26.1% and sand 61.6%) for the andosol and clay loam (clay 16.8%, silt 29.1% and sand 54.1%) for the gray lowland soil. The phosphate adsorption coefficient was 2250 for the former and 1160 for the latter. The other soil properties for the six plots are shown in Table 2. The values of total carbon and nitrogen, cation exchange capacity (CEC), biomass carbon and nitrogen, available nitrogen and FDA hydrolytic activity were smaller in the gray lowland soil than the andosol. The accumulation of total carbon and nitrogen, expressed as the ratios for plots amended with organic fertilizers to those treated with inorganic fertilizers, was almost identical in the two soils. However, the values for the accumulation of biomass carbon and nitrogen were smaller for the gray lowland soil than the andosol.

Changes in Percentage of Pesticides Remaining and Dissipation Rates

Figure 1 shows the changes in the percentage of pesticides remaining in the pots of the two soils treated with inorganic fertilizers. The pesticides were divided into three groups based on dissipation patterns. The first group consists of dimethoate; the decrease in WSF accounted for a large portion of the dissipation, which was rapid. The second group consists of simazine in the andosol and flutolanil; WSF remained stable at a low level, and the dissipation was relatively slow. The dissipation rates for the third group were intermediate values. This group consisted of simazine in the gray lowland soil, and fenobucarb, prometryn and alachlor; WSF abruptly decreased in the first several days of the incubation period, while the corresponding amount of ASF did not decrease in the same period, suggesting a shift of WSF into ASF. For individual pesticides, the other two plots of each soil showed similar dissipation patterns.

The dissipation of the pesticides appeared to follow biphasic profiles throughout the experimental period. The rate constants (k) following the first order kinetics for most pesticides were therefore calculated using the data up to the time when the pesticides remaining decreased to less than

Table 2. Soil properties for the six plots

Plots	pH (H ₂ O)	Total C (%)	Total N (%)	CN	CEC ^{a)} (meq/100 g)	Available P ₂ O ₅ (mg/100 g)	Biomass C (mg/100 g)	Biomass N (mg/100 g)	Available N (mg/100 g)	$\mathrm{B/A^{b)}}$	FDA $(\Delta A_{490}/\mathrm{g} \cdot \mathrm{hr})$	MWHC ^{e)} (%)
A-IF	6.2	8.97	0.56	16.1	37.1	6.9	41.0	5.8	6.1	95.1	0.046	92.8
A-CM	6.7	9.74 (1.09)	0.71 (1.27)	13.7	45.3 (1.22)	15.6 (2.26)	58.6 (1.42)	9.3 (1.60)	12.5 (2.05)	74.1	0.056 (1.22)	98.8 (1.06)
A-CF	0.9	10.02 (1.12)	0.68 (1.21)	14.8	38.1 (1.03)	9.0 (1.30)	56.0 (1.36)	8.3 (1.44)	12.4 (2.03)	67.5	0.066 (1.43)	96.9 (1.04)
G-IF	6.1	1.95	0.17	11.3	16.6	8.4	29.6	5.4	5.6	95.7	0.026	49.6
G-CM	6.3	2.44 (1.25)	0.22 (1.28)	11.1	18.6 (1.12)	27.6 (3.29)	36.7 (1.23)	6.5 (1.21)	7.4 (1.31)	88.1	0.036 (1.38)	51.8 (1.04)
G-SR	0.9	2.08 (1.07)	0.20 (1.14)	10.6	17.4 (1.05)	9.2 (1.10)	37.9 (1.28)	6.9 (1.28)	9.4 (1.67)	73.2	0.031 (1.19)	50.1 (1.01)
a) Cot: 20	00000	a) Cotion and promoter b) The metic of historica M to	is of hismond N	lious of	oblo M o Movim	riogno villalla Maxima lamina holdina	oneocity,					

" Cation exchange capacity. " The ratio of biomass N to available N. " Maximal water holding capacity.

The figure in parentheses denotes the ratio of value for plot amended with organic fertilizers to that for the corresponding inorganic fertilizers-treated plot.

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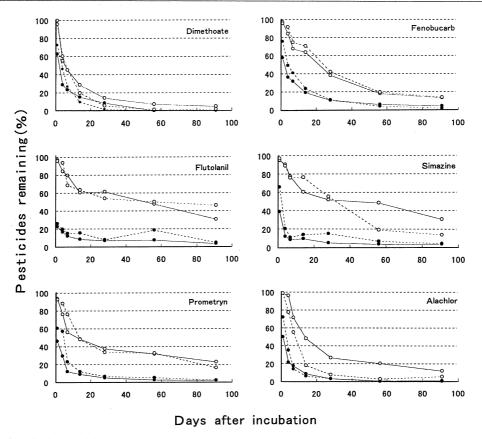


Fig. 1. Changes of pesticides remaining in the plots treated with inorganic fertilizers in the two soils. Symbols denote ○: ASF, ●: WSF, solid line: andosol and dashed line: gray lowland soil.

50% of the amounts added initially. As shown in Table 3, in terms of k values, the ranking was first group \geq third group > second group, the same as for water solubility. This result indicated that the dissipation rates of the pesticides depended on the behavior of WSF. The k values in the gray lowland soil were approximately the same or greater than those in the andosol; the ratio of the average k value for plots in the gray lowland soil to those in the andosol was smallest for fenobucarb at 0.93, followed by flutolanil at 1.08, and largest for simazine at 1.83, followed by alachlor at 1.60. The addition of organic fertilizers to the soil was less effective in stimulating the dissipation of pesticides in the gray lowland soil than the andosol. The ratios of k values for plots amended with organic fertilizer to those for the plots treated with inorganic fertilizers were generally smaller in the gray lowland soil than the andosol; the average values for the six pesticides were A-CM at 1.36, A-CF at 1.39, G-CM at 1.24 and G-SR at 1.12. The $t_{1/2}$ values of the pesticides investigated excluding fenobucarb were smaller than corresponding values in the literature (Table 1).

3. WSF/ASF Ratios of Pesticides

As shown in Table 4, the WSF/ASF ratios of pesticides excluding dimethoate in the gray lowland soil and flutolanil decreased abruptly between one day and four days after

incubation, then gradually. These changes resulted from a rapid decrease in WSF. As discussed above, the dissipation rates depended on the behavior of WSF. Therefore, the data on WSF/ASF ratios at one day after incubation should be used when discussing the relationship between the k values and the ratios. The variation in the ratios at one day after incubation were relatively small for each soil, then increased with incubation time. The ratios were greater in the gray lowland soil than the andosol.

DISCUSSION

The relative standard deviations of rate constants for dissipation and those of soil properties are shown in Table 5. The values of $k_{\rm BC}$, $k_{\rm BN}$, $k_{\rm FDA}$, $k_{\rm TC}$ and $k_{\rm TN}$ for most pesticides excluding flutolanil in the andosol, and simazine, prometryn and alachlor in the gray lowland soil were less variable than the corresponding k values. Indeed, the k values for simazine and prometryn in the gray lowland were extremely small. In contrast, the values of $k_{\rm AN}$ and $k_{\rm AP}$ were more variable than the corresponding k values. Both total carbon and nitrogen were positively correlated with biomass carbon and nitrogen were positively correlated with biomass carbon and nitrogen $^{(1)}$ and FDA hydolytic activity. Elomass nitrogen accounted for a large proportion of available nitrogen. However, the ratios of biomass nitrogen to available nitrogen for the plots amended with organic fertilizers were

Table 3. Rate constants for dissipation and half-life values for the six pesticides

	1	Dimethoate				Fenobucarb				Flutolanil	-	
Plots	k^{a} (day ⁻¹)	t _{1/2} ^{b)} (day)	Period ^{c)} (day)	R^2	$k (\mathrm{day}^{-1})$	<i>t</i> _{1/2} (day)	Period (day)	R^2	$k (day^{-1})$	t _{1/2} (day)	Period (day)	R^2
A-IF	0.089±0.082	7.8	1-14	0.92*	0.030 ± 0.005	23.3	1-56	**66.0	0.008 ± 0.004	88.8	1-91	0.81
A-CM	0.140 ± 0.124 (1.57)	5.0	1-14	0.92*	0.043 ± 0.012 (1.43)	16.2	1-56	**96.0	0.008 ± 0.006 (1.00)	88.8	191	*99.0
A-CF		4.7	1-14	**66.0	0.035 ± 0.004 (1.17)	19.7	1-56	**66.0	0.010 ± 0.005 (1.25)	70.0	1-91	0.82
G-IF	0.120 ± 0.025	5.8	1-14	*86.0	0.029 ± 0.004	23.7	1-56	**66.0	0.008 ± 0.004	87.1	191	0.76*
G-CM	0.169 ± 0.006 (1.40)	4.1	1-14	*86.0	0.037 ± 0.004 (1.28)	18.9	1-56	**66'0	0.011 ± 0.005 (1.38)	63.5	191	**68.0
G-SR	0.164 ± 0.044 (1.37)	4.2	1-14	**66.0	0.035 ± 0.010 (1.21)	20.1	1-56	**96.0	0.009 ± 0.005 (1.12)	78.7	101	0.84**
G/A ^{d)}	1.21				0.93				1.08			
		Simazine				Prometryn				Alachlor		
Plots			Period	, c	4		Period	î			Period	r L
	$k (\mathrm{day}^{-1})$	<i>t</i> _{1/2} (day)	(day)	*	k (day ')	<i>t</i> _{1/2} (day)	(day)	K.	k (day ')	t _{1/2} (day)	(day)	¥
A-IF	0.012±0.009	57.3	1-56	0.77*	0.031 ± 0.023	22.1	1-28	*/8.0	0.051 ± 0.012	13.6	1-28	0.98
A-CM	0.017 ± 0.010 (1.42)	41.0	1–56	0.84*	0.044 ± 0.022 (1.42)	15.9	1-28	0.93**	0.066 ± 0.013 (1.29)	10.5	1-28	0.99**
A-CF	0.020 ± 0.010 (1.67)	34.5	1-56	0.89**	0.032 ± 0.029 (1.03)	22.0	1-28	*08.0	0.079 ± 0.021 (1.55)	8.8	1-28	.*86.0
G-IF	0.029 ± 0.008	24.2	1-56	**96.0	0.040 ± 0.014	17.3	1–28	**96.0	0.098 ± 0.037	7.0	1-28	0.96**
G-CM	0.031 ± 0.010 (1.07)	22.4	1–56	0.95	0.049 ± 0.011 (1.23)	14.3	1-28	**86.0	0.103 ± 0.029 (1.05)	6.7	1-28	**86.0
G-SR	0.031 ± 0.002 (1.07)	22.7	1-56	0.97	0.033 ± 0.009 (0.83)	21.3	1-28	**86.0	0.112 ± 0.039 (1.14)	6.2	1-28	**96'0
G/A	1.83				1.14				1.60			

 $^{\circ}$ k: rate constants for dissipation, predicted values $\pm 95\%$ confidence intervals. The figure in parentheses denotes the ratio of the value for the plot amended with organic fertilizers to that for the corresponding inorganic fertilizers-treated plot. $^{\circ}$ predicted values, $^{\circ}$ period used for calculating the k values following the first order kinetics. R^2 ; regression coefficient for a first order fit, * and **; significantly different at p < 0.05 and p < 0.01, respectively. $^{\circ}$ The ratio of the average k value for plots in the gray lowland soil to that in the andosol.

Table 4.	WSF/ASF	ratios	of	pesticides
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_			And	losol					Gray lov	land soil		
Pesticides	A	Average (%	(o)	Relat	ive standa	rd (%)	A	Average (%	6)	Relat	ive standa	rd (%)
· -	1 d ^{a)}	4d ^{b)}	7d°)	1d	4d	7d	1d	4d	7d	1d	4d	7d
Dimethoate	58.2	48.2	42.0	7.3	6.5	25.3	79.1	80.7	64.9	3.9	7.7	13.3
Fenobucarb	59.6	46.4	42.3	9.8	7.9	10.0	69.7	59.3	58.0	11.3	8.1	5.6
Flutolanil	20.5	19.8	16.2	14.0	17.9	14.0	25.7	22.0	20.7	4.5	21.8	12.7
Simazine	36.9	13.0	11.8	11.1	4.2	7.2	66.6	23.4	19.3	18.6	21.2	43.3
Prometryn	43.6	31.0	25.3	14.9	22.0	41.5	65.9	57.1	41.8	4.2	12.0	25.2
Alachlor	50.7	24.0	23.7	4.1	9.2	10.7	62.7	49.0	20.9	15.3	17.1	21.3

a),b),c) One day, four days and seven days after incubation, respectively.

Table 5. Relative standard deviations of rate constants for dissipation

Soils	Pesticides		Rel	ative standa	ard deviation	ns of rate c	onstants for	dissipation	(%)	
Solis	Pesticides	k	$k_{\scriptscriptstyle m TC}$	$k_{\scriptscriptstyle ext{TN}}$	$k_{\scriptscriptstyle{ ext{CEC}}}$	$k_{ ext{AP}}$	$k_{\scriptscriptstyle \mathrm{BC}}$	$k_{\scriptscriptstyle \mathrm{BN}}$	$k_{\scriptscriptstyle m AN}$	$k_{\scriptscriptstyle ext{FDA}}$
Andosol	Dimethoate	25.1	20.3	15.1	23.1	28.6	9.1	8.1	14.5	12.8
	Fenobucarb	18.3	15.4	8.1	8.8	22.2	8.2	9.9	28.2	17.8
	Flutolanil	13.3	9.8	14.2	19.7	38.8	17.8	22.9	38.1	10.3
	Simazine	26.7	21.1	19.3	28.5	36.3	15.1	16.2	18.9	9.6
	Prometryn	19.9	18.7	13.8	8.5	24.3	16.2	17.9	35.4	23.9
	Alachlor	21.2	16.0	14.0	23.1	34.2	11.2	14.2	23.7	3.
	Average	20.8	17.0	14.1	18.6	30.7	12.9	14.9	26.5	13.0
Gray lowland soil	Dimethoate	17.9	12.4	9.1	13.8	47.1	6.4	7.8	13.6	7.0
	Fenobucarb	11.4	6.0	2.9	6.6	46.6	4.9	5.8	17.7	5.4
	Flutolanil	16.4	4.7	4.6	10.5	42.2	11.6	12.9	22.4	3.
	Simazine	4.1	8.3	8.4	2.7	49.5	9.7	9.1	22.0	12.3
	Prometryn	19.8	14.2	17.2	16.7	45.1	23.5	24.0	34.5	18.8
	Alachlor	6.7	12.2	11.1	7.6	51.5	8.7	7.6	19.4	14.3
	Average	12.7	9.6	8.9	9.6	47.0	10.8	11.2	21.6	10.3

 k_{TC} , k_{TN} , k_{CEC} , k_{AP} , k_{BC} , k_{BN} , k_{AN} and k_{FDA} denote rate constants per total C, total N, CEC, available P₂O₅, biomass C, biomass N, available N and FDA hydrolytic activity, respectively.

smaller than those for the corresponding plots treated with inorganic fertilizers as shown in Table 2. These results indicated that the dissipation rates of most pesticides depended on microbial amount and activity, and related soil properties as reported. ^{5-7,9,11)}

Although the values for microbial amount and activity were smaller in the gray lowland soil than the andosol, the k values in the former were similar to or greater than those in the latter. Except for alachlor, the average k value in the gray lowland soil to that in the andosol (k_G/k_A) was positively correlated with the average WSF/ASF ratio at one day after incubation in the gray lowland soil to that in the andosol [(WSF/ASF)_G/(WSF/ASF)_A] at the 5% level as shown in Fig. 2. As shown in Fig. 3, the values of $k_{\rm BC}$ and k_{BN} in the andosol became less variable with an increase in the WSF/ASF ratio at one day after incubation. The ratio was negatively correlated with the relative standard deviation at the 1% level. This phenomenon was not observed in the gray lowland soil. In the previous report, the leachate concentrations of pesticides ranked in the same order as the corresponding magnitude of the WSF/ASF ratio. 20) Bioavail-

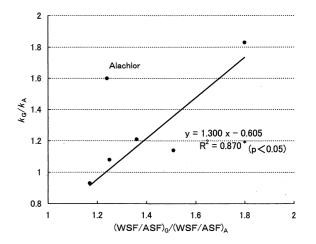
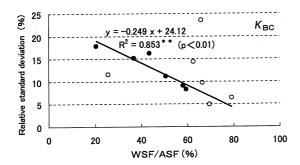


Fig. 2. Relationship between k_G/k_A and (WSF/ASF)_G/(WSF/ASF)_A. k_G/k_A : The average k value in the gray lowland soil to that in the andosol. (WSF/ASF)_G/(WSF/ASF)_A: The average WSF/ASF ratio of pesticides at one day after incubation in the gray lowland soil to that in the andosol.



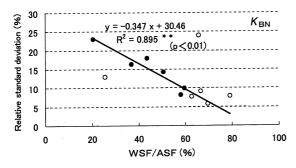


Fig. 3. Relationship between the WSF/ASF ratios of pesticides at one day after incubation and relative standard deviations of $k_{\rm BC}$ and $k_{\rm BN}$. Symbols denote \bullet : and sol and \odot : gray lowland soil. Regression equations are for andosol.

ability refers to the concentration of chemicals in soil solution readily accessible to microorganisms. 23,24) Therefore, the labile fractions of pesticides remaining partially depend on the WSF/ASF ratio.

The applications of organic fertilizers retard the degradation of chemicals as well.²⁵⁾ The variable effects are ascribable to the competing relationship between the degradation and adsorption processes. However, the retardation of dissipation by adsorption was not observed in the present study. Based on the small variation in the WSF/ASF ratio, the inhibition of the dissipation of pesticides would be negligible.

The long-term successive applications of organic fertilizers were less effective in the dissipation of pesticides for the gray lowland soil than the andosol. A possible explanation is that the treatments are less effective in the accumulation of microbial biomass in the gray lowland soil than the andosol, which is ascribable to the small value of CEC for the survival of microorganisms.²⁶⁾

REFERENCES

1) K. Roppongi: Spec. Bull. Saitama Hortic. Exp. Stn. 4, 1-57

- (1995) (in Japanese).
- K. Kamiyama, S. Fujiwara and H. Funahashi: Bull. Agric. Res. Inst. Kanagawa Prefec. 136, 31-42 (1995) (in Japanese).
- M. Katamine, K. Kamewada, Y. Suzuki, Y. Ito and F. Uchida: Bull. Tochigi Agric. Exp. Stn. 50, 79-91 (2001) (in Japanese).
- P. G. Saffigna, D. S. Powlson, P. C. Brookes and G. A. Thomas: Soil Biol. Biochem. 21, 759-765 (1989).
- 5) T. B. Moorman and S. S. Harper: J. Environ. Qual. 18, 302-306 (1989).
- J. V. Pothuluri, T. B. Moorman, D. C. Obenhuber and R. D. Wauchope: J. Environ. Qual. 19, 525-530 (1990).
- T. C. Mueller, T. B. Moorman and C. E. Snipes: J. Agric. Food Chem. 40, 2517-2522 (1992).
- K. Sakamoto and Y. Oba: Soil Sci. Plant Nutr. 37, 387-397 (1991).
- 9) A. S. Felsot and E. K. Dzantor: Environ. Toxicol. Chem. 14, 23-28 (1995).
- 10) T. Murata, H. Tanaka, K. Sakagami, D. Asaka and R. Hamada: Jpn. J. Soil Sci. Plant Nutr. 68, 249-256 (1997) (in Japanese).
- 11) J. P. E. Anderson: Soil Biol. Biochem. 16, 483-489 (1984).
- 12) J. Kanazawa (ed.): "Noyaku no Kankyotokusei to Dokuseidetashu," Godo Publishers, Tokyo, pp.11, 121, 157, 174, 285 and 318, 1996 (in Japanese).
- 13) Agricultural Production Bureau, Ministry of Agriculture, Foresty and Fisheries (ed.): "Nippon no Kochidojo no Jittai to Taisaku," Dojohozen Chosajigyo Zenkokukyogikai, Tokyo, pp. 36-55, 1979 (in Japanese).
- 14) E. D. Vance, P. C. Brookes and D. S. Jenkinson: Soil Biol. Biochem. 19, 703-707 (1987).
- 15) K. Sakamoto and A. Hayashi: Soil Microorganisms 53, 57-62 (1999) (in Japanese).
- 16) A. Hayashi, K. Sakamoto and T. Yoshida: Jpn. J. Soil Sci. Plant Nutr. 68, 322-326 (1997) (in Japanese).
- 17) B. Rotman and B. W. Papermaster: Proc. Natl. Acad. Sci. USA 55, 134-141 (1966).
- 18) B. Lundgren: Oikos 36, 17-22 (1981).
- 19) J. Schnürer and T. Rosswall: Appl. Environ. Microbiol. 43, 1256-1261 (1982).
- 20) S. Suzuki: J. Pestic. Sci. 25, 1-9 (2000).
- 21) G. Guan, T. Marumoto, H. Shindo and M. Nishiyama: Jpn. J. Soil Sci. Plant Nutr. 68, 614-621 (1997) (in Japanese).
- 22) S. Tokuda and M. Hayatsu: Soil Sci. Plant Nutr. 48, 865-869 (2002).
- 23) A. V. Ogram, R. E. Jessup, L. T. Ou and P. S. C. Rao: Appl. Environ. Microbiol. 49, 582-587 (1985).
- 24) W. F. Guerin and S. A. Boyd: Appl. Environ. Microbiol. 58, 1142-1152 (1992).
- 25) J. Rouchaud, F. Gustin, O. Cappelen and D. Mouraux: Bull. Environ. Contam. Toxicol. 52, 568-573 (1994).
- 26) K. Sakamoto and N. Hodono: Soil Sci. Plant Nutr. 46, 483-490 (2000).