

Loss of Amide Herbicides in Runoff Water and Sediment and Their Persistence in Soil

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ABSTRACT

Alachlor and butachlor at the rate of 4.0 kg ai/ha metolachlor at the rate of 3 kg ai/ha, and propanil at the rate of 6 kg ai/ha were applied to the cultivated soil of 8-10 and 25-28% slopes during the middle of rainy season at Royal Angkhang Highland Agriculture Research Station, Fang district, Chiangmai. The soils of the 8-10 and 25-28% slopes were clayloam and clay, respectively. At the 8-10% slope site the maximum concentrations of alachlor, butachlor, metolachlor, and propanil in the runoff were 46.1, 61.5, 65.0 and 86.3 µg/L respectively. At the 25-28% slope site the maximum concentration of alachlor, butachlor, metolachlor, and propanil in the runoff were 18.5, 15.5, 51.4 and 70.3 µg/L, respectively. The highest amounts of water, soil, and herbicides were lost from both sites 1 to 3 days after application. The average amount of alachlor, butachlor, metolachlor, and propanil in the water phase were 66.4, 52.2, 68.0 and 54.9% of the total amount of herbicides found in the combined sediment and water phases, respectively. The maximum concentrations of herbicide were applied to leaves and to sandy loam soil where test plants were grown under greenhouse condition. Plant dry weight were reduced significantly by all herbicides when foliar application was practiced. The plant dry weight were not affect by soil application of herbicides. This indicating that all the chemicals were adsorbed by clay particle and organic matter. Total losses of all herbicides were less than 0.5% of the applied rates. The downward movement of the herbicides to the depth of 7.5-15 cm. was as follows: metolachlor > propanil > alachlor > butachlor which was in correlation with their water solubility. The persistence of alachlor, butachlor, metolachlor, and propanil in the soil of the treated area which could be detected by gas chromatography were 180, 180, 117, and 30 days after application, respectively.

Key words : runoff water, alachlor, butachlor, metolachlor, propanil

INTRODUCTION

A variety of herbicides is used in considerable quantity to control weeds in intensive crop production. Some of these chemicals are applied to plants or soil surface while others are incorporated into the soil. The particular herbicides applied are dictated by crops and weeds. It is possible that some of these chemicals could be transported from the field in runoff waters and pollute other water bodies at lower elevations. Thus, it is important to manage the crop production system (erosion control) to eliminate the pollution hazard while maintaining high productivity.

Alachlor [2-chloro-*N*-(2,6-diethylphenyl)-*N*-(methoxymethyl) acetamide], butachlor [*N*-butoxymethyl)-2-chloro-*N*-(2,6-diethylphenyl) acetamide], metolachlor [2-chloro-*N*-(2-ethyl-6-methylphenyl)-*N*-(2-methoxy-1-methylethyl) acetamide], and propanil [*N*-(3,4-dichlorophenyl) propanamide] are classified as amide family of herbicides. Pudhipreechapong et al (1988) reported that 257,210 kg ai of alachlor, 191,980 kg ai of butachlor, 4,136 kg ai of metolachlor, and 100,361 kg ai of propanil were imported

into Thailand in 1988. Alachlor, metolachlor are used preemergence mainly in corn, soybean, peanut, cotton, sunflower, and certain other ornamentals and established turf species for control of most annual grasses and certain broadleaf weeds (Anonymous, 1989). Butachlor is used preemergence and early postemergence for control of many annual grasses, certain broadleaf weeds and many aquatic weeds in transplant, seeded and upland rice (Anonymous, 1989). Propanil is used for selective postemergence control of annual grasses especially *Echinochloa* spp. in rice (Anonymous, 1989). Eventhough, each year amide herbicides are used in large amount in Thailand but information on transportation of these herbicides in runoff water from agricultural land is not available. This is because the study on herbicide concentration in runoff water is very complicated, time consuming, and costly.

Eventhough, the total amount of herbicide loss in runoff was very small compared to application rate (Wauchope, 1978) the concentration of each single loss should not be dismissed. They might affect susceptible crops growing at the lower elevation or toxic to other aquatic

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organisms where the runoff water get into.

This study did not intend to compare the losses in runoff among the amide herbicides tested because they were applied at different rates. The objectives of this study are to determine; 1. the maximum concentration of amide herbicides in runoff, 2. the total losses of amide herbicides from slopes of different steepness under natural rainfall condition, 3. the toxicity of amide herbicides to sensitive crops when they were applied at maximum concentration detected in runoff, 4. the downward movement of amide herbicides, and 5. the persistence of amide herbicides in soil.

MATERIALS AND METHODS

Field experiment An experiment was conducted at Royal Angkhang Highland Agriculture Research Station, Fang district, Chiangmai. Herbicides were applied on 8-10% and 25-28% slopes where the soil is clay loam (33% sand, 32% silt, 35% clay, 5.12% organic matter, and pH 5.8) and clay (24% sand, 33% silt, 43% clay, 4.76% organic matter, and pH 5.5) respectively. Plot size was 10 m by 2 m and surrounded with 22.5 cm high corrugated roofing sheets to prevent the runoff. The lower edge of the sheets penetrate the soil to 5cm. At the base of the slope a 4.5 cm diameter pipe was connected from the sheet to a 200 liter tank for collecting runoff. The tank was covered to prevent entry of rain. Every day that runoff was observed the water and sediment in the collecting tanks were sampled for extraction.

Herbicides (Table 1) were applied to the soil surface by a motor driven knapsack sprayer with a pressure of 2.72 kg/cm² on August 29, 1983, when the soil moisture was 23%. The main weed was *Imperata cylindrica*. Weed stover residue was cleaned up and the soil surface was cultivated one day before herbicide application. Herbicide was applied on the area of 5 m by 2 m in the upper part of the plot. The treatments were replicated 3 times on each slope in randomized complete block. Glyphosate at 3 kg ai/ha was also applied for weed control in the middle of August.

The volume of runoff water and eroded soil sediment in the collecting tank was measured and thoroughly stirred before randomly collecting for 2.5 L per tank, then the tank was cleaned. Water and sediment samples of 50 ml were extracted 3 times, 150 ml of the solvent listed in Table 1. The other 50 ml sample of water and sediment was filtered through layers of Watman No. 1 and No. 40 filter papers. The sediment was collected, dried, and weighed to determine the amount of eroded soil. The filtrate was extracted 3 times by the solvent listed in Table 1 at the same volume as described above. A preliminary study indicated that the herbicide was completely extracted from the soil and water by the appropriate solvent. After partitioning, the extracting solvents were pooled together and filtered through glass wool and sodium sulfate anhydrous for the separation of sediment and water, then evaporated by a flash evaporator until the volume was reduced to 100 ml. The extracting solvent was transported to the laboratory for herbicide

determination.

At 1, 11, 20, 30, 60, 117, 180 and 241 days after application of herbicides soil samples from 0-7.5 cm and 7.5-15 cm depths were collected from 3 random spots on treated and nontreated areas of each plot with an auger of 2.0 cm diameter. The collected soil was placed in a glass bottle with excess solvent and transported to the laboratory.

Laboratory experiment The volume of solvent from runoff extraction was reduced to 10 ml before gas chromatography.

The analysis of herbicide concentration in soil was done after the solvent was evaporated then soil was ground and passed through 20 mesh screen then dried and weighted. The ground soil was twice extracted by solvent on the shaker (200 cycle/minute) for 20 minutes. The volume of solvent : soil was 3 : 1. The extracting solvents were combined together and the volume reduced to 10 ml before gas chromatography.

The tracer 560 gas chromatography with⁶³ Ni electron capture detector and the column of 1.5% ov-17 + 1.95% ov-202 on Chromosorb WHP 80/100 mech (glass column 2 m long and 0.62 cm width) was used in this study. The oven, detector, and inlet temperatures were 200, 350, and 240°C, respectively. The output and input attenuation were 8 and 10, respectively. N₂ flow and chart speed were 1.12 kg/cm² and 37.5 cm/h, respectively. The result was the average of 3 injections and this was compared to the known standard concentration of each herbicide.

Greenhouse experiment Pregerminated cucumber (Jed-bai), sorghum (KU 439), and rice (R.D.23) were planted in sand and sandy loam soil in 300 ml cups at 10 plants per cup. Five days after planting the plants were thinned to 5 plants per cup. Hoagland solution and water were subirrigated to the cups to keep the soil moisture at field capacity. At 10 days after planting herbicides at the concentration given in Table 4 (base on the maximum concentration detected in runoff) were applied to both sand and soil surfaces for root uptake study. The same concentration of herbicides were applied to leaves of test plants grown in sand culture every 2 days for 3 consecutive applications. During application to leaves the sand surface was covered with sawdust to prevent soil contamination by the herbicides. Herbicides were applied by laboratory sprayer with a 8004 E nozzle, a pressure of 2.04 kg/cm² and spray volume of 500 L/ha. Phytotoxicity of herbicides and the dryweight of plants were recorded at 14 days after the first application of herbicides. Treatments were replicated 4 times in a randomized complete block design.

RESULTS AND DISCUSSION

The maximum concentration of alachlor, butachlor, metolachlor, and propanil found in runoff water and eroded soil sediment were 46.1, 61.5, 65.0 and 86.3 µg/L, respectively from 8-10 slope and 15.5, 15.5, 51.4 and 70.3 µg/L respectively from 25-28% slope (Table 4). Higher amounts of herbicides were detected from 8-10% slope than those of

Table 1 Water solubility and formulation of amide herbicides and solvent used for extracting the herbicides from runoff water and eroded soil sediment.

Herbicides	Water solubility (Anonymous, 1989)	Formulation	Solvent	Rate
	(mg/L)			(kg(ai)/ha)
Alachlor	242 (25°C)	Emulsifiable concentrate	ethyl acetate	4.0
Butachlor	23 (24°C)	Emulsifiable concentrate	ethyl acetate	4.0
Metolachlor	530 (20°C)	Emulsifiable concentrate	xylene	3.0
Propanil	500 (25°C)	Emulsifiable concentrate	xylene	6.0

Table 2 Total volume of runoff water and amount of eroded soil sediment from 8-10 and 25-28% slopes.

Herbicides	8-10% slope			25-28% slope		
	Soil sediment	Water	Sediment: water	Soil sediment	Water	Sediment: water
	(kg/ha)	(L/ha)	(kg:L)	(kg/ha)	(L/ha)	(kg:L)
Alachlor	3,174.3	853,200	1:269	3,255.7	573,700	1:176
Butachlor	3,147.3	853,320	1:271	3,377.4	470,900	1:139
Metolachlor	4,111.5	527,600	1:128	3,911.2	426,000	1:109
Propanil	3,284.3	595,400	1:181	3,301.8	523,505	1:159

Table 3 Total amount of amide herbicides lost in runoff (water phase plus eroded soil sediment phase)

Herbicides	Rate	Total losses in runoff			
		8-10% slope		25-28% slope	
		(g/ha)	(%) ¹	(g/ha)	(%) ¹
Alachlor	(kg ai/ha) 4.0	7.6±2.7	0.19	3.7±1.4	0.09
Butachlor	4.0	15.3±7.7	0.38	2.6±2.1	0.06
Metolachlor	3.0	14.2±2.5	0.47	9.7±1.1	0.32
Propanil	6.0	15.8±2.4	0.26	10.7±2.9	0.18

¹ % of application rate

Table 4 The maximum concentration of amide herbicides detected in water plus eroded soil sediment phases and in water phase.

Herbicides	Rate	8-10% slope		25-28% slope		Average herbicides in water phase
		Water+ sediment	Water	Water + sediment	Water	
		(kg ai/ha)	(µg/L)	(µg/L)	(%)	
Alachlor	4.0	46.1±20.3	31.1±16.5	18.5±9.3	12.1±9.6	66.4
Butachlor	4.0	61.5±20.8	33.1±11.2	15.5±9.3	7.8±6.1	52.2
Metolachlor	3.0	65.0±19.0	46.1±25.7	51.4±2.6	33.5±3.1	68.0
Propanil	6.0	86.3±15.2	46.6±13.0	70.3±7.0	39.2±4.0	54.9

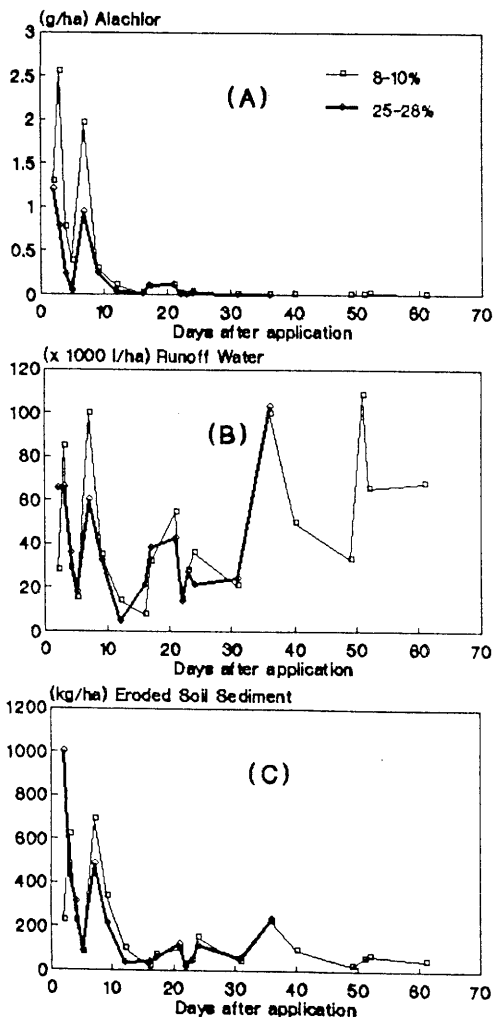


Figure 1 Amount of alachlor in runoff water and soil sediment (A), amount of runoff water (B), and eroded soil sediment (C) from 8 - 10% and 25 - 28% slopes.

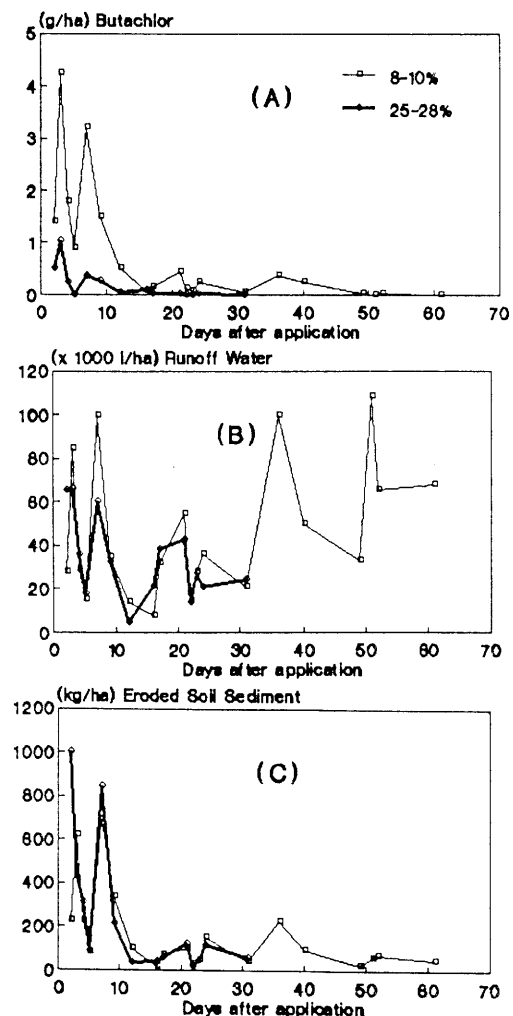


Figure 2 Amount of butachlor in runoff water and soil sediment (A), amount of runoff water (B), and eroded soil sediment (C) from 8 - 10% and 25 - 28% slopes.

25-28% slope because runoff water and eroded soil sediment was lost from 8-10% slope in greater amount. Furthermore, the soil of the 8-10% slope was clayloam which might be susceptible to erosion than clay of 25-28% slope. The maximum concentration of herbicides in runoff was observed at 2 to 3 days after application when high amount of runoff and soil loss occurred (Figure 1 to 4). After these periods, most of herbicide concentration in runoff were drastically decreased. The concentration of herbicides in runoff were correlated with herbicide concentration at the soil surface (Leonard et al, 1979). Amide herbicides residue on soil surface are most subject to rapid volatilization, photodecomposition, microbial degradation, and leaching. In this study halflives of alachlor, butachlor, and propanil in soil of both slopes were ranged from 5 to 10 days and metolachlor from 5 to 15 days (Figure 5 and 7). However

herbicide concentrations in runoff might increase when high amount of runoff water and eroded soil were observed (Figure 1 to 4).

Alachlor, butachlor, and metolachlor at 46, 62, and 65 $\mu\text{g/L}$ (maximum concentration detected in runoff, Table 4) reduced test plant dry weight when applied to leaves and to root of plants grown in sand culture (Table 5). Propanil at 86 $\mu\text{g/L}$ reduced test plant dry weight when applied to sand culture (Table 5). However, dry weight of test plants was not reduced when the herbicides at 3 fold above concentration were applied to sandy loam soil except alachlor (Table 5). These results indicated that all herbicides were absorbed by the leaves and were toxic to test plants. Furthermore, when they were applied to sandy loam soil they might be adsorbed by clay particle and organic matter. Since all amides reduced test plant dry weight when applied to leaves

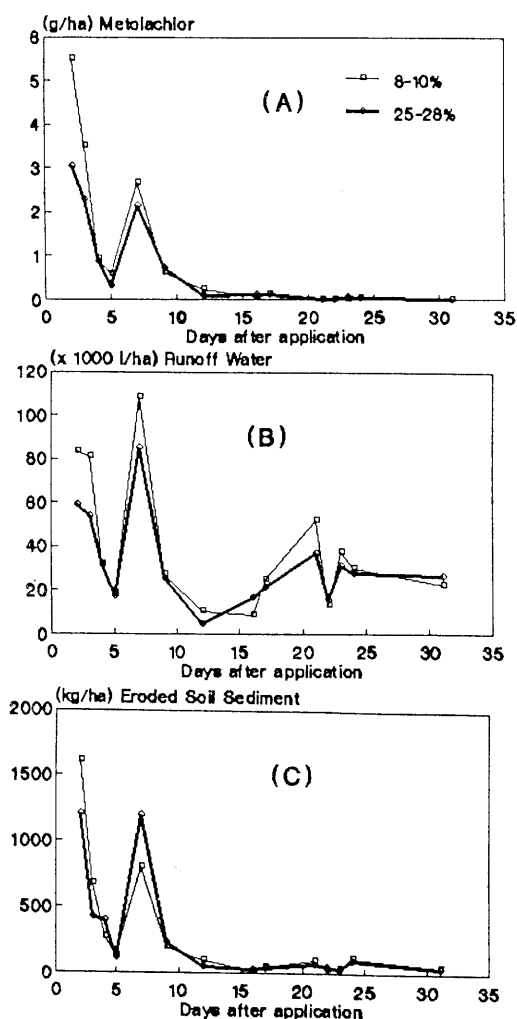


Figure 3 Amount of metolachlor in runoff water and soil sediment (A), amount of runoff water (B), and eroded soil sediment (C) from 8 - 10% and 25 - 28% slopes.

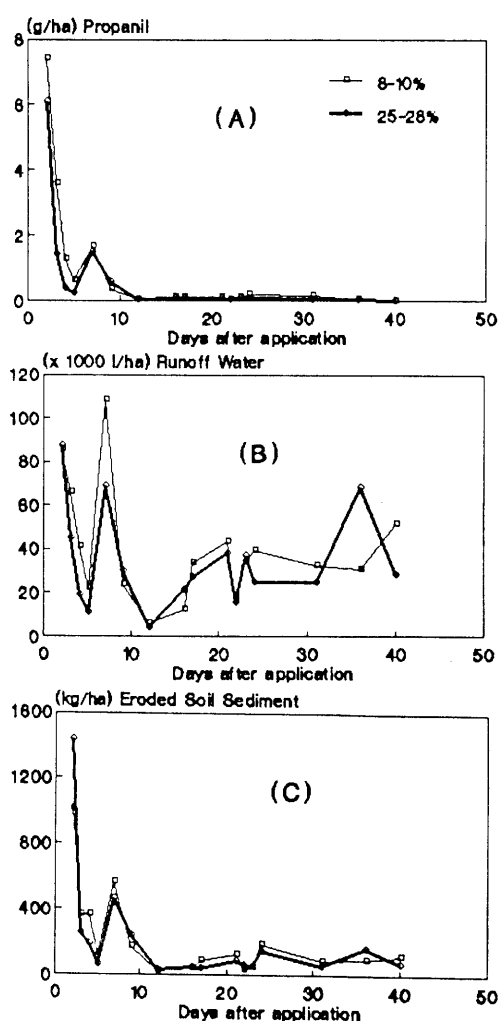


Figure 4 Amount of propanil in runoff water and soil sediment (A), amount of runoff water (B), and eroded soil sediment (C) from 8 - 10% and 25 - 28% slopes.

(Table 5), the use of runoff water which transported directly from application area to leaves of susceptible crops must be avoided. However in our study in the field, distance between the border of treated area and collecting tanks was only 5 m. The concentration of amide herbicides in runoff water might be reduced by the degradation process of aerobic microorganism (Pionke and Chesters, 1973) where the treated area was far beyond. The susceptible upland crops were used as test plants in this study. The susceptibility of other aquatic organisms to amides at concentration detected in runoff should be studied. There was a report that accumulation of alachlor and dinoseb residue and toxicity of alachlor do not present great environmental risks to fish, but the toxicity of dinoseb may pose risk (Call et al, 1984).

Alachlor, butachlor, metolachlor, and propanil detected in the water phase of the runoff were 66.4, 52.2, 68.0

and 54.9%, respectively (Table 4). The solubility of amides in water are in the order, metolachlor > propanil > alachlor > butachlor (Table 1). In other studies, 60% (Wu, 1980) and 68% (Correll et al, 1979) of alachlor were found in water phase. The amount of butachlor found in soil sediment was more than other amides (Table 4). Although propanil was high soluble in water (Table 1) its mobility in soil was very little (Kearney, 1977). The concentration of herbicides in water phase were not only relatively correlated with their water solubility but also their adsorbability and the properties of adsorbents (Wauchope, 1978). Paraquat and arsenicals (Wauchope, 1978) including glyphosate (Sprankle et al, 1975) are high water soluble herbicides but are strongly adsorbed by soil particle, therefore most of them will be in the sediment phase. As discussed previously all amides in this study was adsorbed onto clay mineral and organic

Table 5 Effect of amide herbicides on testing plants.

Herbicides	Concentration ⁴	Applied to root		Applied to leaves ⁶
		sand	soil ⁵	
	(ug/L)		(g/plant)	
Alachlor ¹	0.0	0.036	0.48	0.071
	46.0	0.032	0.45	0.053
	92.0	0.030	0.43	0.042
	138.0	0.027	0.42	0.038
	LSD _{.05}	0.003	0.06	0.008
	LSD _{.01}	0.004	NS	0.011
	C.V. (%)	5.00	4.10	9.19
Butachlor ²	0.0	0.114	0.80	0.499
	62.0	0.101	0.70	0.298
	124.0	0.094	0.89	0.236
	186.0	0.078	0.87	0.210
	LSD _{.05}	0.012	NS	0.058
	LSD _{.01}	0.018	NS	0.083
	C.V. (%)	8.01	24.7	11.62
Metolachlor ²	0.0	0.209	0.96	0.277
	65.0	0.173	1.07	0.251
	130.0	0.144	1.42	0.229
	195.0	0.127	1.74	0.185
	LSD _{.05}	0.031	NS	0.022
	LSD _{.01}	0.045	NS	0.031
	C.V. (%)	12.03	29.9	8.26
Propanil ³	0.0	0.185	1.06	0.334
	86.0	0.160	0.85	0.296
	172.0	0.143	1.19	0.255
	258.0	0.125	1.23	0.240
	LSD _{.05}	0.028	NS	0.026
	LSD _{.01}	0.040	NS	0.037
	C.V. (%)	11.23	11.8	6.84

1 Rice (R.D. 23)

2 Sorghum (KU 439)

3 Cucumber (Jed-bai)

4 The maximum concentration of amide herbicides detected in runoff water.

5 Soil was sandy loam (67% sand, 22% silt, 11% clay, and 0.77% organic matter)

6 Herbicides were applied to leaves of testing plant, every 2 days, 3 times.

matter. However, amides are nonionic (Wauchope, 1978) and formulated as emulsifiable concentrate (Table 1). The adsorption of most organic pesticides were highly correlated with soil organic matter content. Herbicide molecules in runoff were in the adsorbed state might be desorbed depending on their concentration in the equilibrium solution. Irreversible adsorption has been observed for monuron in soil and 2, 4-D and amiben in muck soil (Pionke and Chesters, 1973).

The total amount of alachlor, butachlor, metolachlor, and propanil in runoff from 8-10% slope were 7.6, 15.3, 14.2 and 15.8 g/ha which equivalent to 0.19, 0.38, 0.47 and 0.26% of applied rate, respectively and from 25-28% slope were 3.7, 2.6, 9.7 and 10.7 g/ha which equivalent to 0.09, 0.06, 0.32 and 0.18% of applied rate, respectively (Table 3). The total losses of herbicides from 8-10% slope were greater than total losses from 25-28% slope. The total runoff water and eroded soil sediment of 8-10% slope were

lost in greater amount than those of 25-28% slope (Table 2). In addition, the soil of 8-10% slope was loam which might be susceptible to erosion than clay of those 25-28% slope. Many factors might affected herbicide losses in runoff such as the type of soil and topography (Frere, 1975). The losses

of herbicides in this study will be reduced if herbicides were not applied on cultivated soil and at the middle of the rainy season. The crops did not plant in herbicide application area, because crop canopy can reduce soil erosion and herbicide losses (Asmussen et al, 1977). Furthermore,

Table 6 Persistence of herbicides in soil (of treated area).

Herbicides	Rate	8 - 10% slope			25 - 28% slope		
	(kg ai/ha)	(days) ¹	(g/ha)	(%) ²	(days) ¹	(g/ha)	(%) ²
At the level of 0.0-7.5 cm							
Alachlor	4.0	117	2.0±0.001	0.05	180	5.0±0.002	0.16
Butachlor	4.0	180	10.0±0.001	0.24	180	4.0±0.01	0.10
Metolachlor	3.0	117	86.0±0.01	2.73	117	22.0±0.02	0.73
Propanil	6.0	30	23.0±0.03	0.38	30	51.0±0.001	0.83
At the level of 7.5-15 cm							
Alachlor	4.0	117	8.0±0.01	0.19	60	13.0±0.02	0.32
Butachlor	4.0	30	64.0±0.02	1.55	20	4.0±0.01	0.10
Metolachlor	3.0	60	75.0±0.07	2.38	60	130±0.12	4.35
Propanil	6.0	30	60.0±0.10	0.98	30	149±0.012	2.42

1 Days after application of herbicides until the last day which herbicides can be detected.

2 % of applied rate.

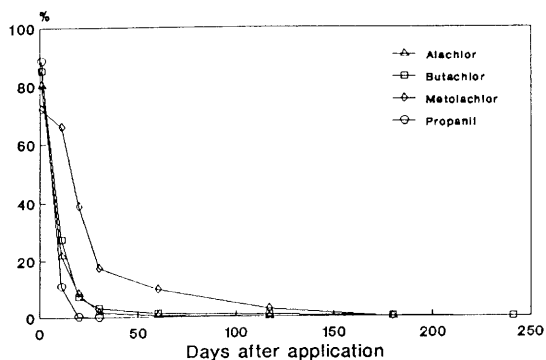


Figure 5 Percentage of herbicides recovered in soil at 0 to 7.5 cm. of 8 - 10% slope.

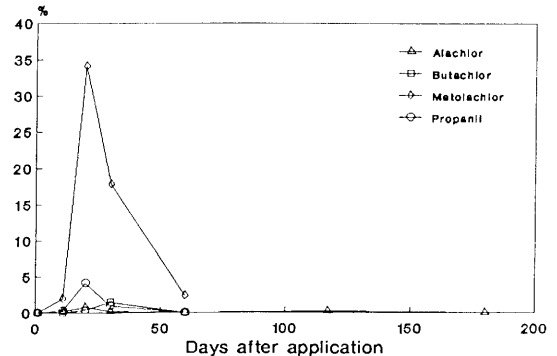


Figure 6 Percentage of herbicides recovered in soil at 7.5 to 15 cm. of 8 - 10% slope.

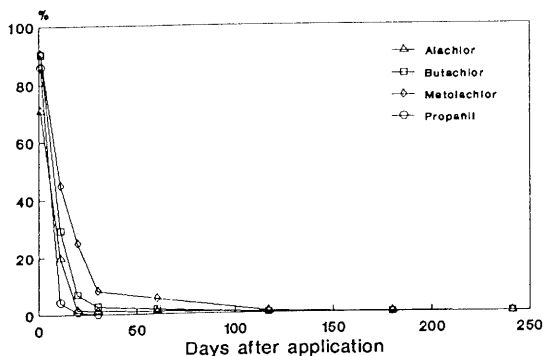


Figure 7 Percentage of herbicides recovered in soil at 0 to 7.5 cm. of 25 - 28% slope.

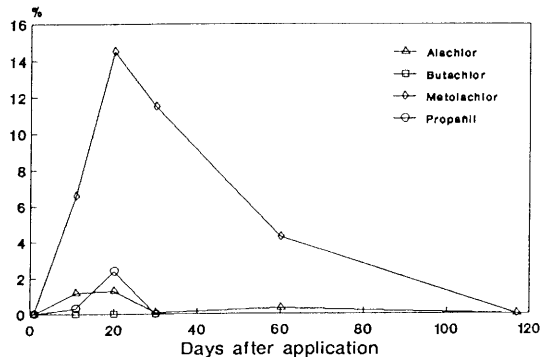


Figure 8 Percentage of herbicides recovered in soil at 7.5 to 15 cm. of 25 - 28% slope.

herbicide losses were reduced when they were applied in no tillage system (Hall et al, 1984). Moreover, the amount of herbicide losses in runoff will be prevented by application of herbicides at the beginning of the rainy season when brief rainfall might occur and increased herbicide infiltration into deep soil before the runoff event would occur (Hall et al, 1983). Hall et al (1984) suggested that planting legumes between the corn rows might reduce soil erosion and herbicide losses in runoff. Moreover, they said the small grain strip at slope base could also reduce soil and herbicide losses. However, herbicides with solubility of 10 ppm or higher are lost mainly in water phase of runoff and erosion control practices will have little effect on such losses (Wauchope, 1978).

The last detectable of alachlor, butachlor, metolachlor, and propanil in runoff from both slopes were at 52, 52, 24 and 36 days after application respectively (Figure 1 to 4). The persistence of alachlor, butachlor, metolachlor, and propanil were 180, 180, 117 and 30 days (Table 6). The duration of losses in runoff correlated with persistence in soil except those of metolachlor. The percentage of metolachlor detected in soil at 11 to 117 days after application was higher than other herbicides (Figure 5 to 8). Since metolachlor is more water soluble it may move downward into the soil (Figure 6). This prevents the chemical from losses by runoff. Furthermore, metolachlor might tolerated to be metabolized by soil microorganisms, or soil microorganisms which could metabolize this herbicide were not available in treated area. By using bioassay method Steward et al (1975) reported that alachlor and propanil persisted in soil for 40 to 70 and 1 to 3 days, respectively. Furthermore, butachlor and metolachlor have not been reported to persist in soil of Thailand and caused phytotoxicity to the following susceptible crops. The amount of herbicides detected in soil at the last days by chromatography method (Figure 5 to 8) might be the bound residues which soil microorganism could not metabolized. As stressed before, loss of amide herbicides from soil surface can be due to evaporation, photodecomposition, and leaching. However, when they were vertically moved into soil they might be subjected to metabolize by various soil microorganisms. High soil moisture after application of herbicides in this study might increase microbial activity.

All herbicides could not be detected in soil of the adjacent (nontreated area) slope base. This is possibly due to the amount of herbicides in runoff transported to those areas were too low to be detected or they might be subjected to degrade by soil microorganisms. The vertical movement of all herbicides was observed at 11 days after application and the maximum concentration, in the 7.5-15 cm depth occurred at 20 days after application on both slopes (Figure 6 and 8). The order of vertical movement of herbicides in soil of both slopes were metolachlor > alachlor > propanil > butachlor (Figure 6 and 8) which were relatively correlated with their water solubility (Table 1). Alachlor and propanil were classified as medium and least movement herbicides, respectively (Kearney, 1977). Beestman and

Deming (1976) reported that dissipation of butachlor in soil was not due to leaching into subsoil. In contrast to butachlor, the leaching of metolachlor into 15 cm was observed in siltloam soil within 18 days with the rainfall of 3.74 to 3.76 cm (Braverman, 1984). Since all herbicides were applied at rather high rate during the middle of rainy season, large amounts of herbicides were detected in the subsoil.

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