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Transport of Triazine Herbicides in Runoff from a Hillside

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Abstract

The triazine herbicides were applied to the cultivated clay loam and clay soils of 8 – 10 and 25 – 28% slopes, respectively at the rate 4 kg (ai)/ha in the middle of the rainy season at Royal Angkhang Highland Agriculture Research Station, Fang District, Chiangmai. High amounts of

herbicides were lost from both slopes 1 to 3 days after application but the loss from 8 – 10% slope were higher. At 8 – 10% slope the maximum concentrations in the runoff were atrazine 64, cyanazine 19, simazine 247, and metribuzin 99 µg/l. At 25 – 28% slope the maximum concentrations in the runoff were atrazine 67, cyanazine 29, simazine 378, and metribuzin 70 µg/l. The average

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amounts in water phase were atrazine 43, cyanazine 68, simazine 46, and metribuzin 72% of the total amount of herbicides found in both sediment and water phases.

The maximum concentration of herbicides were applied to leaves and to sandy loam soil where test plants were grown under greenhouse conditions. No herbicide reduced plant dry weight when applied to leaves indicating that the herbicides were not taken up by leaves. Atrazine, cyanazine, and simazine did not reduce plant dry weight when applied to soil. This is believed to be due to the adsorption of herbicides onto soil colloids. However metribuzin reduced plant dry weight when applied to sandy loam soil indicating that

metribuzin needs soil with higher organic matter and clay to act as adsorbents.

Total loss of atrazine, cyanazine, and metribuzin on the slopes were less than 1% of applied rate respectively while simazine was lost at less than 4%. All herbicides were leached to 7.5 to 15 cm depth at 20 days after application. The herbicides also move laterally on the soil surface to adjacent nontreated slope base. Their persistence in soils were equal to those of the treated area. Persistence of atrazine, cyanazine, simazine, and metribuzin in soil of treated area were 117 days on 8 – 10% slope and 180 days on 25 – 28% slope, respectively.

Key words : triazine, runoff of herbicides

Atrazine (6-chloro-N-ethyl-N'-(1-methylethyl) 1,3,5-triazine-2,4-diamine), cyanazine (2-((4-chloro-6-(ethylamino)-1,3,5-triazine-2-yl) amino)-2-methylpropane nitrile), simazine (6-chloro-N, N'-diethyl-1,3,5-triazine-2,4-diamine) are a symmetrical chlorotriazines while metribuzin (4-amino-6-(1,1-dimethylethyl)-3-(methylthio)-1,2,4-triazine-5(4H)-one) is an asymmetrical triazine. The triazine family are important herbicides for crop production in Thailand; 1,082,324 kg (ai) of atrazine and 12,600 kg(ai) of metribuzin were imported in 1988 (Pudhipreechabong *et al.*, 1988). Atrazine, cyanazine, and simazine are used mainly in corn, sorghum, sugarcane and pineapple; but simazine is also used in orchards and in strawberries. Metribuzin is used in soybean, potatoes, tomatoes, sugarcane and asparagus. Information on triazines loss in runoff are available in other countries but not for Thailand. Studies of the amount

of triazine herbicides in runoff should be conducted to determine method to prevent the contamination of herbicides in water and then effect on the ecosystem.

The amount of runoff losses of herbicides from fields is a complex function. Four factors independent of individual field characteristics and which effect and may even dominate herbicide losses are (i) the physical-chemical properties of herbicides as deposited by an application; (ii) the location of the spray deposit after application (foliage or soil surface or subsurface soil); (iii) the amount of applied on area basis; and (iv) dissipation and degradation of spray deposit before runoff occurs (Wauchope and Leonard, 1980).

Various methods have been applied to reduce atrazine, cyanazine, and simazine loss in the runoff. These methods include planting other crops at the base of slopes (Hall *et al.*, 1983), planting crops with no tillage (Triplett *et al.*, 1978), growing crops as living

and dead mulch between rows of the main crop (Hall *et al.*, 1984), and incorporation of herbicides into soil (Baker and Laflen, 1979).

The objectives of this experiment were to determine (i) the maximum concentration of triazines in runoff; (ii) the total losses of triazines from different slopes under natural rainfall conditions; (iii) the toxicity of triazines to sensitive crops when they were applied at the maximum concentration detected in runoff; (iv) the lateral movement of triazines to surface base of slopes; and (v) the persistence of triazines 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 × 2m and surrounded with 22.5 cm high zinc plate sheets to prevent the runoff. The lower edge of zinc plate penetrate the soil to 5 cm. At the base of the slope a 4.5 cm diameter pipe was connected from the zinc plate 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 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% (Table 1). The main weed was *Imperata cylindrica* (L.) Raeuschel. 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 × 2 m. in the upper part of the plot. The treatments were replicated 3 times on each slope in a randomized complete block. Glyphosate at 3 kg (ai)/ha was also applied for weed control in the middle of August.

Table 1. Water solubility, formulation and applied quantity of triazine herbicides and solvent used for extracting the herbicides from runoff water and soil sediment.

Herbicides	Water solubility (Anonymous, 1989)	Formulation	Solvent	Quantity of applied (kg ai/ha)
	(mg/l)			
Atrazine	33 (27°C)	wettable powder	ethyl acetate	4
Cyanazine	171 (25°C)	wettable powder	benzene	4
Simazine	3.5 (20°C)	wettable powder	acetone	4
Metribuzin	1,220 (20°C)	wettable powder	acetone	4

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 by 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 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 collection soil was placed in a glass bottle with excess solvent and transported to the laboratory.

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	Soil sediment : water	Soil sediment	Water	Soil sediment : water
	(kg/ha)	(l/ha)	(kg:l)	(kg/ha)	(l/ha)	(kg:l)
Atrazine	2,767	604,500	1:260	3,099	471,000	1:177
Cyanazine	2,628	449,400	1:185	1,565	277,500	1:177
Simazine	3,732	793,300	1:212	3,187	551,400	1:173
Metribuzin	3,651	721,100	1:197	3,164	514,400	1:163

Table 3. Total amount of triazine herbicides losses in runoff (water plus eroded soil sediment).

Herbicides	Rate	Total losses in runoff			
		8 – 10% slope		25 – 28% slope	
	(kg ai/ha)	(g/ha)	(%) *	(g/ha)	(%) *
Atrazine	4	24 ± 9	0.60	13 ± 5	0.35
Cyanazine	4	9 ± 3	0.24	5 ± 2	0.14
Simazine	4	115 ± 42	2.89	60 ± 29	1.52
Metribuzin	4	33 ± 6	0.84	12 ± 3	0.30

* % of application rate.

Laboratory experiment

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

For soil analysis 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 analyzing by gas chromatography.

Greenhouse experiment

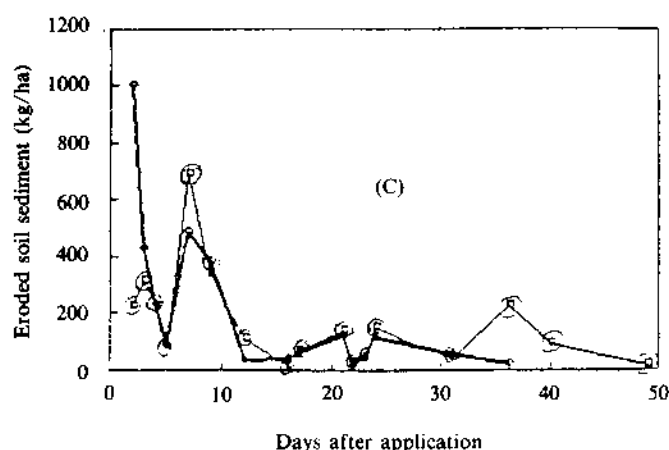
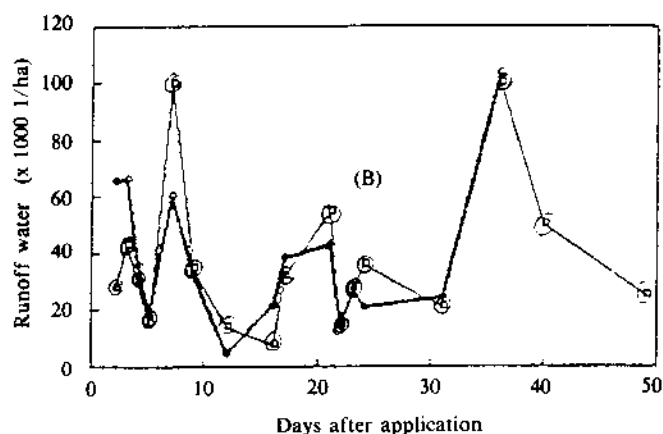
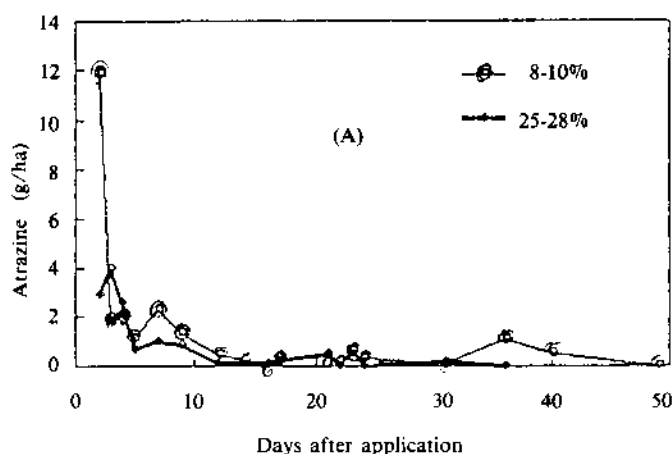
Pregerminated cucumber (Jed – bai) and soybean (S J 5) were planted in sand and sandy loam soil in 300 ml pots at 10 plants per pot. Five days after planting the plants were thinned to 5 plants per pot. Hoagland solution and water were subirrigated to the pots to keep the soil moisture at field capacity. 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 randomized complete block design.

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

Herbicides	Rate (kg ai/ha)	8 - 10% slope		25 - 28% slope		Average herbicides in water (%)
		Water + sediment	Water	Water + sediment	Water	
Atrazine	4	64 ± 14	21 ± 11	67 ± 12	35 ± 15	43
Cyanazine	4	19 ± 4	14 ± 7	29 ± 8	18 ± 6	68
Simazine	4	247 ± 83	113 ± 42	378 ± 89	176 ± 69	46
Metribuzin	4	99 ± 15	61 ± 27	70 ± 14	57 ± 11	72

Results and Discussion

The maximum concentrations of atrazine, cyanazine, simazine, and metribuzin in runoff water and eroded soil sediment are given in Table 4. The concentration of herbicides in runoff were observed to decline 1 to 3 days after application (Figures 1 to 4), then increased at day 7 probably because of high amount of runoff water and eroded soil observed at this period (Figures 1 to 4). The increased loss herbicides at day 7 might be due to their upward movement. Atrazine, simazine (Helling, 1970), and metribuzin (Ladlie *et al.*, 1976) were reported to move upward on TLC plate. Cyanazine moved upward equal to or more than atrazine because of its higher water solubility (Table 1).



(A) Atrazine (B) Runoff water (C) Eroded soil Sediment
Figure 1. Amount of atrazine in runoff water and soil sediment (A), amount of runoff water (B), and eroded soil sediment (C) from 8 to 10% and 25 to 28% slopes.

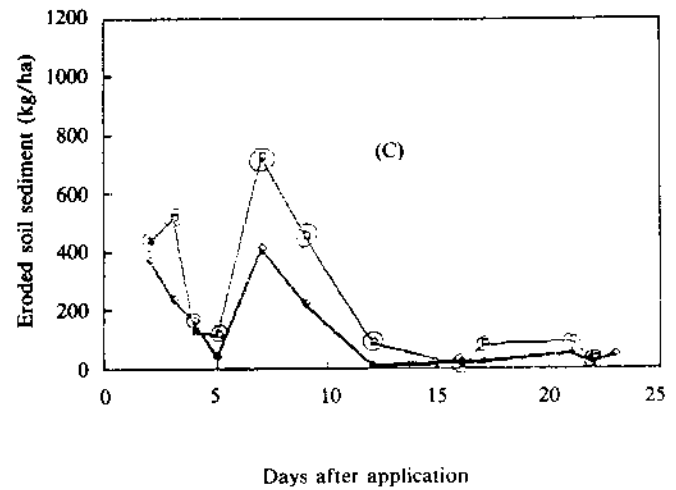
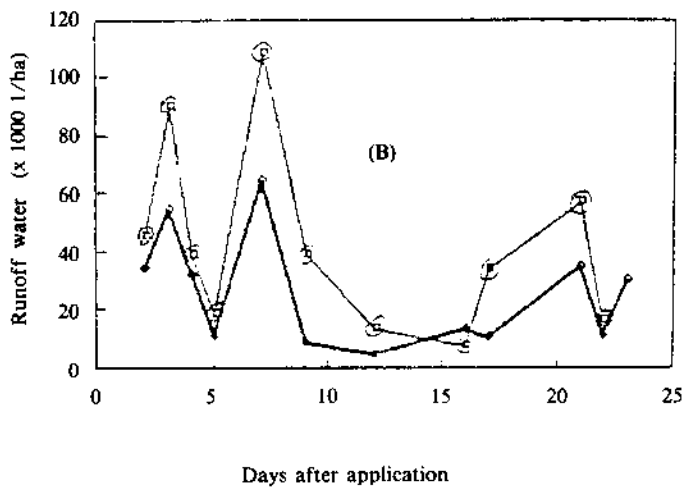
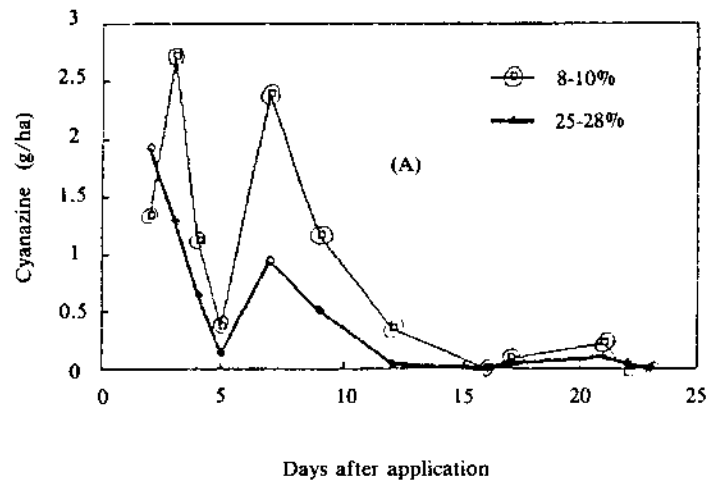


Figure 2. Amount of cyanazine in runoff water and soil sediment (A), amount of runoff water (B), and eroded soil sediment (C) from 8 to 10% and 25 to 28% slopes.

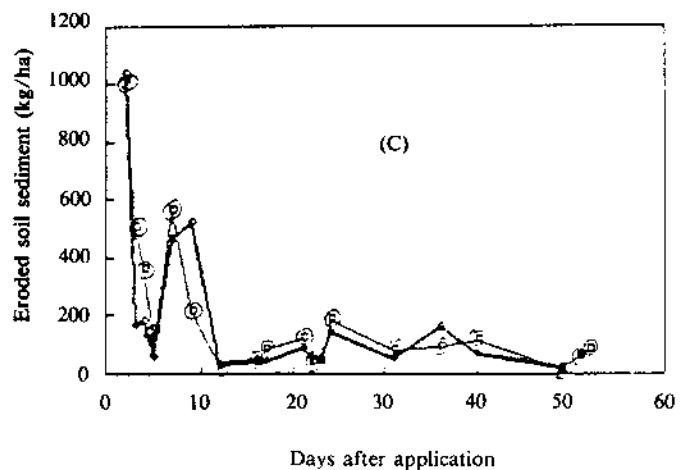
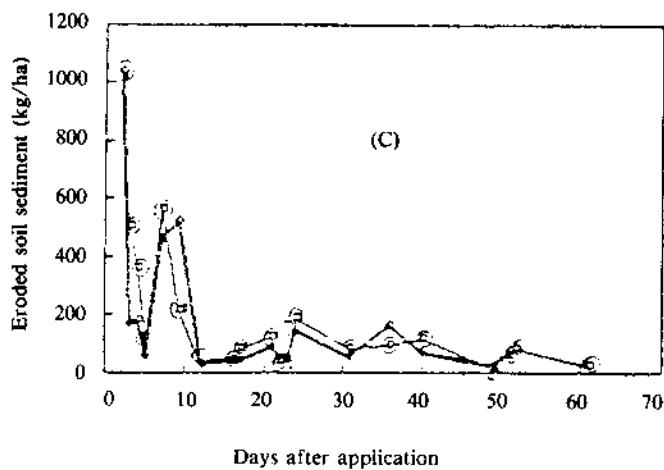
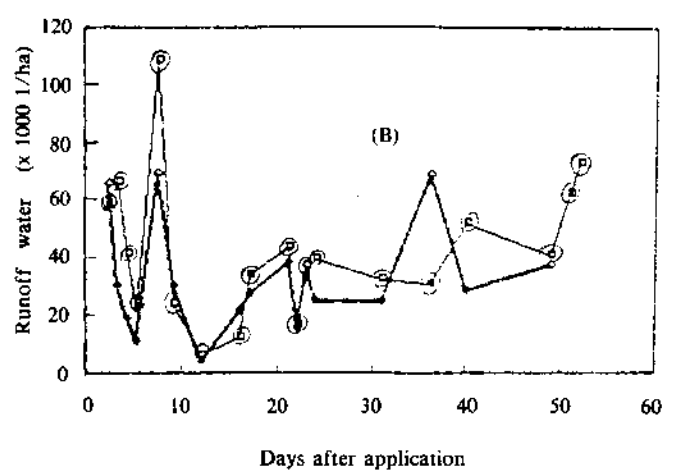
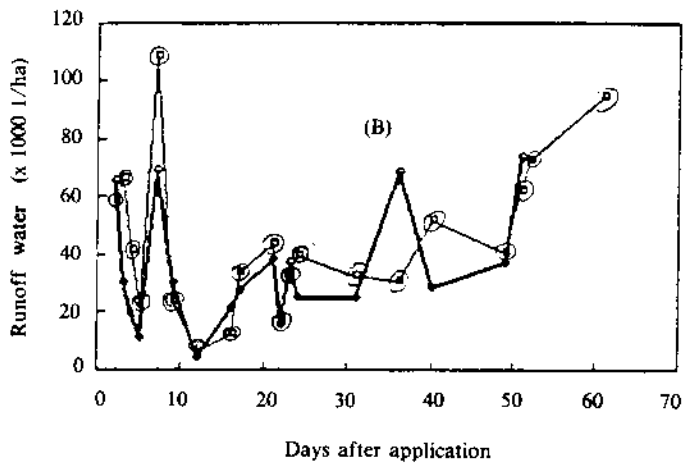
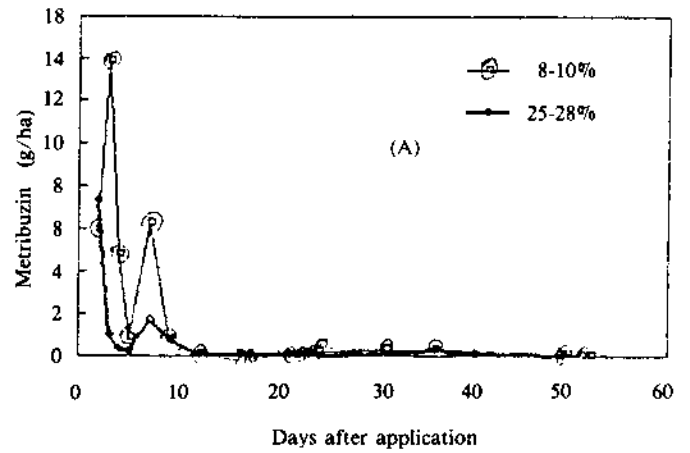
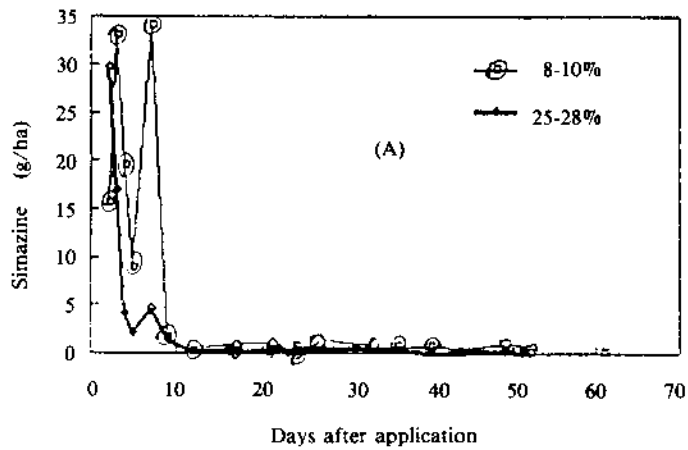


Figure 3. Amount of simazine in runoff water and soil sediment (A), amount of runoff water (B), and eroded soil sediment (C) from 8 to 10% and 25 to 28% slopes.

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

Cyanazine applied at three times the concentration detected in runoff did not reduce plant dry weight when applied to roots of plants grown in soil or sand culture (Table 5). Atrazine, simazine, and metribuzin at 125, 250, and 100 $\mu\text{g/l}$ (the maximum concentrations detected in runoff), respectively reduced plant dry weight where they were applied to the soil surface plants grown

Table 5. Effect of atrazine, cyanazine and simazine on dry weight of SJ 5 soybean.

Herbicides	Concentration	Applied to root		Applied to leaves*
		sand*	soil [®]	
	($\mu\text{g/l}$)		(g/plant)	
Atrazine	0.0	0.229	0.490	0.274
	0.125	0.241	0.400	0.276
	0.250	0.165	0.380	0.297
	0.375	0.132	0.340	0.288
	L.S.D. _{0.05}	0.027	0.140	NS
	LSD _{0.01}	0.038	NS	NS
	C.V. (%)	11.31	23.50	13.98
Cyanazine	0	0.276	0.470	0.302
	30	0.213	0.410	0.298
	60	0.227	0.400	0.268
	90	0.214	0.510	0.322
	L.S.D. _{0.05}	NS	NS	NS
	L.S.D. _{0.01}	NS	NS	NS
	C.V. (%)	22.36	26.50	15.59
Simazine	0	0.259	0.440	0.277
	250	0.196	0.400	0.282
	500	0.141	0.430	0.305
	750	0.123	0.360	0.298
	L.S.D. _{0.05}	0.043	NS	NS
	L.S.D. _{0.01}	0.059	NS	NS
	C.V. (%)	16.15	14.60	11.26

★ Plants were harvested at 10 days after application.

© Plants were harvested at 30 days after application and soil was sandy loam (67% sand, 22% silt, 11% clay and 0.77% organic matter).

Table 6. Effect of metribuzin on dry weight of Jed – bai cucumber.

Herbicides	Concentration	Applied to root		Applied to leaves*
		sand*	soil [®]	
	($\mu\text{g/l}$)		(g/plant)	
Metribuzin	0.0	0.300	0.200	0.650
	100.0	0.016	0.110	0.736
	200.0	0.021	0.030	0.581
	300.0	0.013	0.030	0.540
	L.S.D. _{0.05}	0.034	0.070	NS
	L.S.D. _{0.01}	0.047	0.140	NS
	C.V. (%)	23.54	32.0	21.66

★ Plants were harvested at 10 days after application.

© Plants were harvested at 30 days after application and soil was sandy loam (67% sand, 22% silt, 11% clay and 0.77% organic matter)

on sand culture (Tables 5 and 6). However, when the same concentration of these herbicides were applied to sandy loam soil, only metribuzin reduced plant dry weight (Tables 5 and 6), suggesting that all herbicides except metribuzin were adsorbed in sandy loam soil. When all herbicides were applied to leaves of plants at three times the concentration detected in the runoff plant dry weight were not affected. These suggested that the runoff water from atrazine, cyanazine, and simazine treated areas might be used for irrigation in soybean field with sandy loam soil. However, the rate of application, soil type, and rate of rainfall must be similar to the location in this study since herbicide runoff is site specific (Wauchope and De Doursey, 1989). Ladlie *et al.* (1976) reported that metribuzin was adsorbed in soil at less amount than atrazine. In our experiment other herbicides were adsorbed in sandy loam soil with 11% clay and 0.77% organic matter, but metribuzin, at the concentration of 100g/l

was not adsorbed by this soil. It might be adsorbed onto soil with higher clay and organic matter content. Furthermore the rate of metribuzin used in this study was 4 to 8 fold higher than normal rate. Application at normal rate might reduce herbicide losses in runoff, as herbicides losses in runoff are correlated with their concentration at the soil surface (Leonard *et al.*, 1979).

The concentration of herbicides detected in the runoff in this experiment might be reduced if they were applied under the following conditions.

(i) at normal rate, because the amount of herbicide found in runoff depends on the rate of application (Hall *et al.*, 1972);

(ii) soil surface is undisturbed before application, because non-tilled soil reduces runoff erosion and herbicide movement (Hall *et al.*, 1984);

(iii) application at the beginning of the rainy season when soil has low moisture content, since herbicide runoff did not occur when rainfall intensity did not exceed the soil infiltration rate (Bailey *et al.*, 1974; Wauchope, 1978);

(iv) the duration from application time until the first runoff event should be longer than a few days because the concentration of triazine herbicides in soil will be reduced by biological and nonbiological processes (Jordan *et al.*, 1970; Kaufman and Kearney, 1970) and the concentration of herbicide in the runoff were correlated with concentration at the soil surface (Leonard *et al.*, 1979);

(v) crops were planted in treated area, because a crop canopy can reduce soil erosion and the herbicide losses (Asmussen *et al.*, 1977; Bruce *et al.*, 1975), furthermore growing crops or weeds on the low slopes might reduced the losses of water, soil, atrazine (Hall *et al.*, 1983) and cyanazine (Hall *et al.*, 1984).

The amount of cyanazine and metribuzin found

in water alone were 68 and 72% of total herbicide found in the combined water and sediments, while atrazine was 43 and simazine was 46% (Table 4). The water solubility of cyanazine and metribuzin is higher than those of atrazine and simazine (Table 1), but their adsorbability and properties of adsorbent (clay particle and organic matter) also affect their accumulation in water. Furthermore adsorption and desorption between herbicide and adsorbent might occur during the transportation of runoff (Wauchope and Myers, 1985). Atrazine, cyanazine, and simazine are chloro-s-triazine and their adsorption is highly related to organic matter content. Simazine was reported to be adsorbed onto soil equal or more than atrazine (Weber, 1971). Cyanazine might be adsorbed less than atrazine and simazine due to its higher water solubility (Table 1). Metribuzin as an *as* triazine is adsorbed onto soil less than atrazine because of differences in water solubility and basicity (Ladlie *et al.*, 1976).

The total amount of atrazine, cyanazine, simazine, and metribuzin loss from both slopes are given in Table 3. Furthermore, the total runoff water and eroded soil sediment loss from both slopes are given in Table 2.

The last detections of atrazine, cyanazine, simazine, and metribuzin in runoff were 20, 22, 52, and 51 days after application (Figures 1 to 4). The duration of losses in runoff were not correlated with the persistence of herbicides in soil. The persistence of herbicides were 117 days on 8–10% slope and 180 days on 25–28% slope (Table 7). However, at that period the amount of herbicides remaining in soil were very low as compared with the application rates. At 0–7.5 and 7.5–15 cm levels of both slopes cyanazine and metribuzin were detected less than 1% of the applied rates, but atrazine and simazine were found less than 5 and 10% of the applied rates, respectively.

Table 7. Persistence of triazine herbicides in soil of treated area.

Herbicides	Rate	8 – 10% slope			25 – 28% slope		
		(kg ai/ha)	(days) [★]	(g/ha)	(%) [⊙]	(days) [★]	(g/ha)
At the level of 0.0 – 7.5 cm.							
Atrazine	4	117	160	3.83	180	130	3.07
Cyanazine	4	117	40	0.96	180	3	0.07
Simazine	4	117	160	3.97	180	130	3.10
Metribuzin	4	117	40	0.68	180	5	0.14
L.S.D. _{0.05}			60	1.47		70	1.78
L.S.D. _{0.01}			90	2.22		100	2.69
C.V. (%)			22.6	19.76		20.0	22.01
At the level of 7.5 – 15 cm							
Atrazine	4	117	180	4.19	117	70	1.74
Cyanazine	4	117	30	0.79	117	30	0.57
Simazine	4	117	50	1.21	117	390	9.37
Metribuzin	4	117	30	0.74	117	10	0.35
L.S.D. _{0.05}			60	1.42		170	3.78
L.S.D. _{0.01}			90	2.16		25	5.75
C.V. (%)			20.3	22.91		19.3	17.94

★ Days after application.

⊙ % of applied rate.

The amount of herbicides in the nontreated (slope base) areas were less than those of the treated areas at the same period (Tables 7 and 8). In temperate country, persistence of atrazine was 300–500 days, simazine 200–400 days and metribuzin 150–200 days (Stewart *et al.*, 1975). Therefore persistence of *s*-triazine herbicides in soil were shorter in this study as it was a subtropical area. Hyzak and Zimdahl (1974) reported that metribuzin persisted in soil in shorter periods under high temperature. At the same period atrazine, cyanazine, and simazine were detected in soil at higher amounts than metribuzin (Table 7). This might be due

to different molecular structure. Cyanazine was found to rapidly disappear from soil with the formation of a derivative lacking an isopropyl group as a major phytotoxic metabolite. Furthermore cyanazine was found to degrade more quickly than atrazine. (Kearney, 1977).

Table 8. Persistence of triazine herbicides in soil of nontreated area (slope base).

Herbicides	Rate	8–10% slope			25–28% slope		
		(kg ai/ha)	(days) [★]	(g/ha)	(%) [⊙]	(days) [★]	(g/ha)
At the level of 0.0–7.5 cm							
Atrazine	4	117	30	0.65	177	30	0.69
Cyanazine	4	117	10	0.30	117	8	0.21
Simazine	4	117	20	0.48	117	60	1.54
Metribuzin	4	117	10	0.33	117	20	0.44
L.S.D. _{0.05}			NS	NS		20	0.55
L.S.D. _{0.01}			NS	NS		40	0.84
C.V. (%)			22.6	21.81		31.80	35.45
At the level of 7.5–15 cm							
Atrazine	4	60	3	0.07	60	7	0.18
Cyanazine	4	60	20	0.44	60	20	0.59
Simazine	4	60	2	0.06	60	3	0.07
Metribuzin	4	60	10	0.35	60	5	1.23
L.S.D. _{0.05}			5	0.13		10	0.33
L.S.D. _{0.01}			8	0.19		20	0.50
C.V. (%)			27.0	27.15		28	31.77

★ Days after application.

⊙ % of applied rate.

All herbicides move laterally in runoff on soil surface to an adjacent (nontreated area) slope base, however, they did not persist in the soil of the lower slope longer than those of treated areas (Tables 7 and 8). At 50 days after application, less than 20% of herbicides were found in soil at 0–7.5 cm. level (Figures 5 and 7).

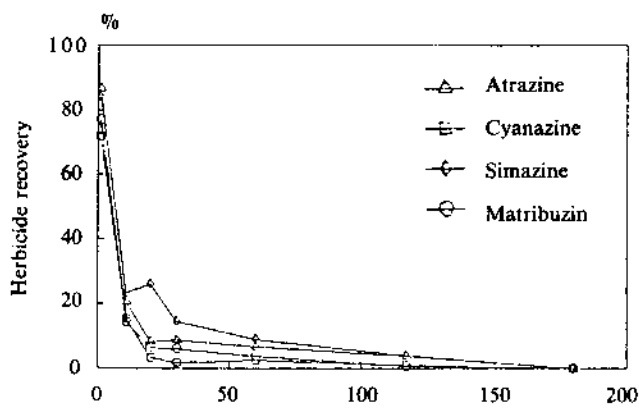


Figure 5. Percentage of herbicides recovered in soil at 0 to 7.5 cm from soil surface of 8-10% slope.

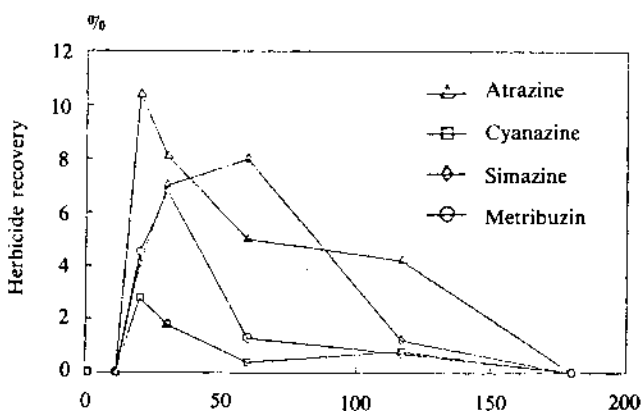


Figure 6. Percentage of herbicides recovered in soil at 7.5 to 15.0 cm from soil surface of 8-10% slope.

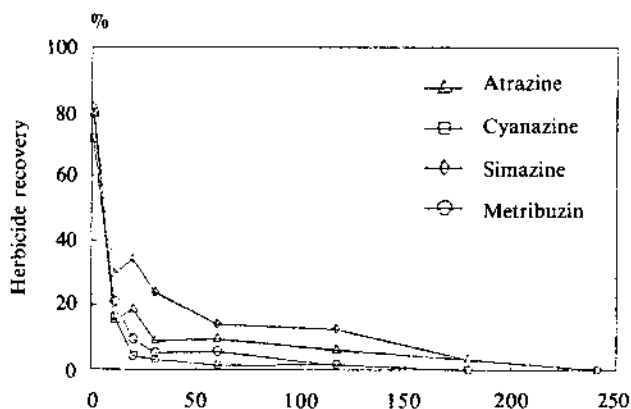


Figure 7. Percentage of herbicides recovered in soil at 0 to 7.5 cm from soil surface of 25-28% slope.

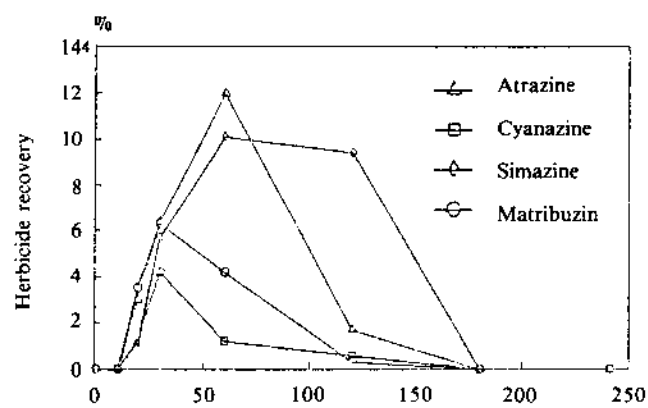


Figure 8. Percentage of herbicides recovered in soil at 7.5 to 15.0 cm from soil surface of 25-28% slope.

Vertical movement of all herbicides to 7.5 – 15 cm level was observed at 20 days after application (Figures 6 and 8). Atrazine and simazine were classified as of moderate mobility in soil while acid and cationic herbicides were very mobile and essentially immobile, respectively (Helling, 1971). Hartly (1964) found that atrazine moved downward more than simazine due to differences in their water solubility. Therefore in *s*-triazine subfamily, cyanazine might moved downward more than atrazine and simazine. However, metribuzin as an *as*-triazines was reported to move in soil more than atrazine due to differences in water solubility, basicity, and steric effects (Ladlie *et al.*, 1976).

Conclusion

Total atrazine, cyanazine, and metribuzin transported from both slopes were less than 1% of applied rate but the total simazine transported was less than 4%. The maximum concentration of atrazine, cyanazine and simazine in the runoff did not reduce soybean dry weight. This indicates that runoff water from these herbicides treated area can be used for soybean irrigation

system. The runoff water from metribuzin treated area might caused injury to susceptible crops especially cucumber, when it was used as irrigated water. However, metribuzin was applied at very high rate (8 fold of normal rate) in this experiment. If metribuzin was applied at normal rates the phytotoxicity of metribuzin from runoff water should not occur. All herbicides move both vertically to deep soil and laterally to slope base.

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