Fate and Effectiveness of Tebuthiuron Applied to a Rangeland Watershed[1]

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ABSTRACT

Tebuthiuron[3] (*N*-[5-(1,1-Dimethylethyl)-1,3,4-triadiazol-2-yl]-*N*,*N*′-dimethylurea), a herbicide, was applied at a rate of 0.84 kg/ha to a rangeland watershed and test plots in pelleted and dissolved forms, respectively. The removal of tebuthiuron in the runoff water from the watershed was determined for each runoff event. After 21 months, total tebuthiuron removed in runoff water was 0.47% of that applied. The low removal rate was credited to the timing of the tebuthiuron application. Tebuthiuron applied to test plots moved to a depth of 15 cm in 8 months with 326 mm of precipitation. Total tebuthiuron remaining in the test plots after 21 months was 38% of the amount applied. The time required for the concentration of tebuthiuron in the test plots to reach non-detectable levels was estimated to be from 2.9 to 7.2 years. Tebuthiuron was effective on the dominant species on the watershed 21 months after treatment, producing an apparent 100% kill for tarbush (*Flourensia cernua* DC.), fourwing saltbush [*Atriplex canescens* (Pursh) Nutt.], and littleleaf sumac (*Rhus microphylla* Engelm. ex Gray), and 61, 92, and 0% for creosote-bush [*Larrea tridentata* (DC.) Cov.], whitethorn (*Acacia constricta* Benth.) and graythorn (*Condalia spathulata* Gray), respectively.

Additional Index Words: **herbicide, brush control, water quality.**

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Over the last 70 to 100 yr, vast areas of the southwestern U.S. rangeland have been invaded by undesirable woody plants. Herbicides have been used to economically control different woody plants on rangelands with considerable success (Bovey et al., 1981; Meyer & Bovey, 1980; Morton et al., 1978). Herbicides that are more effective on woody plants than on grasses at low application rates can greatly improve forage grass quality and establishment on rangelands (Evers, 1981; Bjerregaard et al., 1979; Sosebee, 1979; Baur, 1978; Scifres & Mutz, 1978). The herbicide tebuthiuron[3] (*N*-[5-(1,1-Dimethylethyl)-1,3,4-thiadiazol-2-yl]-*N*,*N*′-dimethylurea) was first used for weed control in industrial areas where total vegetation control was desired (Bjerregaard et al., 1979). Further research has shown that tebuthiuron is effective on many woody plants at lower application rates while significantly increasing forage

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[3] This paper reports results of research only. Mention of a herbicide in this paper does not constitute a recommendation by the USDA, nor does it imply registration under FIFRA. Mention of trademarks or proprietary products does not constitute a guarantee or warranty of the product by the USDA and does not imply their approval to the exclusion of other products that may also be suitable. Trademark names are used only for the information of the reader.

production 2 and 3 yr after application (Jacoby et al., 1982; Scifres et al., 1981; Bjerregaard et al., 1979).

Information on the fate of tebuthiuron applied to rangeland for brush control is limited. Bovey et al. (1978) measured decreasing concentrations of tebuthiuron ranging from 12.0 to 1.4 mg/L in the runoff water from a test plot receiving 38 mm of simulated rainfall during 1 h immediately after application of tebuthiuron as a wettable powder at a rate of 1.12 kg/ha. They also measured an average concentration of 2.23 mg/L during the first runoff event from a 1.3-ha watershed that received 28 mm of natural rainfall, which occurred 2 d after tebuthiuron was applied in pellet form at 2.24 kg/ha.

Tebuthiuron movement to a depth of 61 cm after 6 months has been reported in a Houston Black clay (Udic Pellusterts), typical of the soils in the Blackland Prairie (Bovey et al., 1978). Laboratory investigations have shown that as the clay and organic matter content of a soil increased, the sorption of tebuthiuron increased and its movement decreased (Weber, 1980; Chang & Stritzke, 1977). Soil pH is important in that adsorption of tebuthiuron decreases as pH increases (Weber, 1980). Bovey et al. (1982) measured the persistence of tebuthiuron under field test conditions in a soil and found activity, based on a bioassay technique, after 2.25 yr. These findings of Bovey et al. (1978, 1982), Chang and Stritzke (1977), and Weber (1980) indicate that tebuthiuron used for brush control, especially in the southwestern USA where coarse-textured and low organic matter soils are predominate, could be persistent and a potential nonpoint pollutant in streamflow.

The objectives of this study were to determine: (i) the amount of tebuthiuron removed from a rangeland watershed in the runoff water and adsorbed on sediment, (ii) the depth of tebuthiuron movement in a soil, (iii) the amount of tebuthiuron remaining in a soil profile over time after a surface application, and (iv) the effectiveness of tebuthiuron in killing woody plants on a rangeland watershed.

MATERIALS AND METHODS

The study area used was a 0.34 ha rangeland watershed at an elevation of 1370 m, located on the Walnut Gulch Experimental Watershed near Tombstone, AZ and covered primarily with creosote-bush [*Larrea tridentata* (DC.) Cov.], tarbush (*Flourensia cernua* DC.), whitethorn (*Acacia constricta* Benth.), with small amounts of graythorn (*Condalia spathulata* Gray), fourwing saltbush [*Atriplex canescens* (Pursh) Nutt.], and littleleaf sumac (*Rhus microphylla* Engelm. ex Gray). Two similar soils on the watershed were Rillito and Laveen (Typic Calciorthids). Both soils have a loamy-skeletal particle-size class in the top 30 cm of the profile, with a range of 46 to 71% rock fragments > 2 mm. The particle-size analysis of the fine-earth fraction (i.e., < 2 mm) for both soils was about 60, 26, and 15% for sand, silt, and clay-sized fractions, respectively. The topographic slope of the watershed varied from 5 to 10%. Mean monthly temperatures at the site for January through December were 8.8, 10.0, 12.2, 16.4, 21.1, 25.6, 26.3, 25.1, 23.7, 18.9, 12.9, and 9.0°C, respectively. A 29-yr mean monthly precipitation at the site for January through December was 20.0, 13.4, 14.7, 7.9, 3.0, 12.4, 93.2, 83.6, 29.2, 15.0, 10.4, and 19.1 mm, respectively. Class A pan evaporation at the site, on a yearly basis, averaged 2588 mm.

Adjacent to the watershed, two replicate 3 by 3 m test plots were constructed by sieving the top 60 cm of soil to remove particles > 13 mm. The modified soil in the test plots than had 48% of mineral particles > 2 mm in size. The < 2 mm fraction was characterized as 63, 20, and 17% sand, silt, and clay, respectively. Soil bulk density, 10 months after disturbance, was determined to be 1.60 Mg/m^3. Test plot preparation in this manner was necessary to allow uniform core sampling of the soil with depth without the impediment caused by the rock fragments in the soil profile. The organic C in the test plots was 1.05% (Allison, 1965) and the slope was 3%.

On 3 Feb. 1981 the watershed was hand-treated with tebuthiuron at a rate of 0.84 kg/ha active ingredient in 0.32-cm diameter extruded pellets. The test plots were hand-sprayed at the same time and rate with tebuthiuron dissolved in an ethanol-water mixture. The "liquid" tebuthiuron was used to achieve a uniform surface application to allow for the determination of the actual amount of tebuthiuron applied, amount remaining with time, and depth of movement through the use of soil core sampling.

Precipitation at the study area was measured by a recording raingauge. An H-flume, with a continuous water-level recorder, was used to measure the surface runoff at the watershed outlet. A pump sampler located at the H-flume removed aliquots of water and suspended sediment at 3-min intervals throughout each runoff event, and deposited them into individual sample bottles. Sampling throughout the flow depth was accomplished by a pivoting tube from the flume floor with a float on the end and equidistant holes along the tube. Bedload sediment samples were collected from deposition in the drop box of the H-flume after each runoff event.

After the summer precipitation, 4 m-wide transects were made across the watershed, and for each creosote-bush, tarbush, and whitethorn encountered, percent defoliation and viability or kill were determined visually. Plants on the watershed of lesser dominance were evaluated individually for herbicidal effects.

Four 5.7-cm diameter soil core samples were taken to a depth of 7.5 cm before and after the tebuthiuron was applied to the test plots. The soil cores were sectioned into two increments at 2.5 cm from the soil surface, and each of the four increments of 2.5 and 5.0 cm from a test plot were composited to make a complete sample. The purpose of the initial soil core sampling was to determine if there were any residues in the soil that would give a false indication of tebuthiuron and the exact amount applied. Four soil core samples from each test plot were also taken on selected dates and composited into depth increments. All water and suspended sediment, bedload sediment, and soil samples were kept refrigerated until analysis. Analysis of the water and suspended sediment samples represents both tebuthiuron in the soluble form in the runoff water and sorbed to the suspended sediment. Methods of extraction and analysis of tebuthiuron used in this study have been described by Loh et al. (1980). The detection limits were 1.0 μg/L for the water and suspended sediment and 0.05 mg/kg for the bedload sediment and soil samples.

RESULTS AND DISCUSSION

The amount of precipitation available for leaching tebuthiuron into the soil before 8 July 1981, the first significant runoff event after tebuthiuron application, was 86 mm. Tebuthiuron concentration in the runoff water from the 8 July event was the highest observed, and ranged from a high of 91 μg/L to a low of 40 μg/L (Fig. 1). During the 8 July runoff event, the tebuthiuron concentration increased, decreased, and finally remained constant with time, closely mimicking the runoff hydrograph. The next runoff event, on 12 July, started in the same pattern, but was of short duration, so only two samples were obtained. An explanation of the concentration pattern with time, seen in the 8 July and at the start of the 12 July events, could be from a higher concentration of tebuthiuron near the soil surface and within the effective depth for extraction into the surface runoff water. The higher tebuthiuron concentration near the soil surface probably resulted because the rainfall following tebuthiuron appliation was insufficient to leach the tebuthiuron below the effective depth for extraction. Starting with the 19 July event, the tebuthiu-

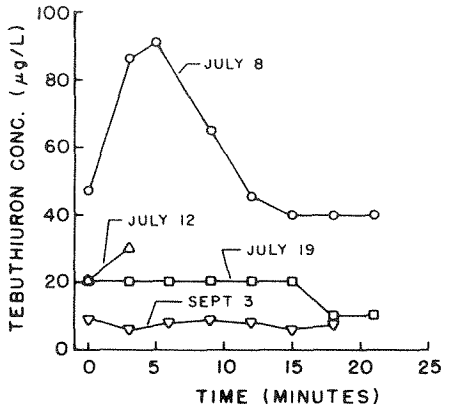

Fig. 1—Tebuthiuron concentration in runoff water and suspended sediment from the watershed with time for selected events in 1981.

ron concentration pattern in the runoff became essentially constant or decreasing with time during each storm event (Fig. 1). This pattern continued through the 1982 runoff events. During this time, the last sample collected from a runoff event usually had a lower concentration than the first sample of the next event. The solubility of tebuthiuron in water is 2300 mg/L, and it was reasonable to conclude that the herbicide was moving into the soil with infiltration, hence a generally lower concentration in the runoff water with time for each event and from one event to the next. Soluble tebuthiuron was probably returning to the soil surface by upward capillary movement as surface evaporation took place between runoff events and as a result, produced the higher concentrations at the beginning of subsequent runoff events. By the last runoff event of 1982, the concentration in the runoff water was at, or below the detection limit, indicating that the tebuthiuron was depleted from the effective depth of interaction with surface runoff. Tebuthiuron depletion was attributed to leaching to a deeper depth and microbial decomposition. Evidence for surface depletion was found in the test plots, as no detectable tebuthiuron residue was found in the top 2.5 cm after the 1982 runoff season (Table 2).

Total tebuthiuron removed in the water and suspended sediment for each event was calculated as a product of the concentration and discharge integrated over the hydrograph by the trapezoidal rule (Table 1). The total amount of tebuthiuron applied to the watershed was 289 g, and the amount removed in runoff water and suspended sediment over all events represented 0.42 and 0.049% for 1981 and 1982, respectively. Total tebuthiuron removed from the watershed in the bedload sediment was extremely small, and not considered a significant pathway for removal from the watershed. The bedload sediments had a sandy textural classification, and were associated with tebuthiuron concentrations in the μg/L range in the runoff waters. Chang and Stritzke (1977) studied sorption of tebuthiuron on a Eufaula sand (Psammetic Paleustalfs) and found that, for an equilibrium solution concentration of 1 mg/L, about 0.2 μg of tebuthiuron was sorbed per gram of soil. Therefore, the small amount of tebuthiuron observed in the bedload sediment is consistent with their results.

Table 1—Tebuthiuron removed, runoff volume, and precipitation for each runoff event from the watershed during 1981 and 1982.

Date	Removal form: Suspended sediment and soluble†	Removal form: Bedload sediment	Runoff volume	Storm precipitation‡
	mg		L	mm
		1981		
30 June	2.6	0.01	80	12.7
8 July	328.6	0.09	6 360	20.3
12 July	26.8	0.07	1 090	6.4
19 July	161.6	0.11	9 300	21.6
21 July	5.2	NDR§	460	6.4
25 July	3.4	NDR	310	9.7
28 July	100.7	NDR	7 740	25.4
30 July	125.7	NDR	11 950	14.0
30 July	72.6	NDR	13 230	20.3
1 Aug.	27.6	NDR	3 350	6.4
10 Aug.	196.4	NDR	26 590	20.8
12 Aug.	11.2	NDR	1 380	5.6
13 Aug.	6.5	NDR	910	5.6
19 Aug.	8.0	NDR	950	10.2
31 Aug.	84.9	2.86	16 790	17.3
3 Sept.	63.6	0.41	8 440	14.5
Totals	1 225.4	3.55	108 930	216.9
		1982		
6 July	3.48	0.34	6 650	24.9
19 July	1.57	NDR	1 650	5.8
20 July	2.13	NDR	4 540	17.3
30 July	1.74	NDR	2 180	8.4
31 July	1.60	NDR	1 930	7.6
1 Aug.	0.23	NDR	260	2.8
3 Aug.	4.04	0.02	1 300	6.4
11 Aug.	0.67	0.11	730	7.4
12 Aug.	12.73	NDR	4 870	7.1
23 Aug.	46.09	NDR	34 070	30.5
10 Sept.	37.29	NDR	18 610	22.9
11 Sept.	30.38	NDR	42 020	21.1
11 Sept.	0.34	NDR	850	8.1
Totals	142.3	0.47	119 660	170.3

† Suspended sediment and soluble = tebuthiuron sorbed to suspended sediment and soluble in runoff water.
‡ Storm precipitation = rainfall water associated with each runoff event.
§ NDR = no detectable residue.

Linear regression was used to examine relationships between the storm precipitation, runoff volume, and tebuthiuron removed with the suspended sediment and water for each runoff event (Table 1). No significant relationship was found between the storm precipitation and runoff volume, but the r^2 value for tebuthiuron removal vs. runoff volume for 1982 was 0.78. The lack of a significant relationship between the same parameters in 1981 was due primarily to the large removals of the tebuthiuron remaining at the soil surface during the first few runoff events. As the tebuthiuron was depleted in the soil surface in 1982, a much higher storm intensity and runoff volume was required to remove the tebuthiuron, and this tended to produce the linear relationship between runoff volume and tebuthiuron removal. Ahuja and Lehman (1983) suggested that the transfer of a chemical from below a thin soil surface layer may be due to the pumping action of turbulence generated by raindrop impact and may be considered as an accelerated-diffusion process.

Tebuthiuron was applied to the test plots at a calculated rate of 0.84 kg/ha. However, analysis of the soil after application revealed an actual rate of 0.63 kg/ha (Table 2). The disparity was attributed to spray drift

Table 2—Concentration of tebuthiuron in soil with depth over time in test plots.

Sample date	Depth (cm) 0-2.5	2.5-7.5	7.5-15	15-23	23-30.5	Total
	kg/ha					
3 Feb. 1981						
Plot I	0.62	NDR†				0.62
Plot II	0.64	NDR				0.64
Control I‡	NDR	NDR				
Control II‡	NDR	NDR				
20 July 1981						
Plot I	0.11	0.33	NDR			0.44
Plot II	0.11	0.36	NDR			0.47
8 Oct. 1981						
Plot I	0.08	0.22	0.08	NDR	NDR	0.38
Plot II	0.07	0.18	0.08	NDR	NDR	0.33
16 June 1982						
Plot I	0.01	0.21	0.15	NDR	NDR	0.37
Plot II	0.01	0.22	0.11	NDR	NDR	0.34
27 Oct. 1982						
Plot I	NDR	0.12	0.11	NDR	NDR	0.23
Plot II	NDR	0.14	0.11	NDR	NDR	0.25

† NDR = no detectable residue.
‡ Control I and Control II = samples taken before tebuthiuron application.

and application outside the plot areas, since the only vegetation within 1 m of the plots, a creosote-bush, showed symptoms of tebuthiuron injury. Eight months after application, tebuthiruon had moved to a depth of 15 cm, with 326 mm of precipitation. The majority of this movement was suspected to have taken place during the months of July through September, when over 50% of the annual precipitation occurs. Leaching to a deeper depth was not observed in 1982, with an additional 260 mm of precipitation (Tables 2 and 3). Bovey et al. (1978) applied tebuthiuron to test plots as a wettable powder and pellets at a rate of 2.24 kg/ha, and reported movement to a depth of 61 cm following 353 mm of precipitation in a 3-month period. The last sampling in our study indicated that tebuthiuron had been removed from the top 2.5 cm by surface runoff, leaching to a deeper depth and/or decomposition. Thirty-eight percent of the applied tebuthiuron remained in the profile after 21 months, indicating that tebuthiuron can be persistent in soil, at least under the conditions of this study.

Even though the soil in the test plots was modified, the amount of tebuthiuron removed in surface runoff from the test plots was probably as small as it was on the watershed. Therefore, most of the decrease in the total amount of tebuthiuron in the soil profile was attributed to decomposition. Tebuthiuron decomposition rate varied tremendously, as measured by the slope of a line that would connect the data points on Fig. 2. The slow rate between 247 and 497 d was attributed to low soil moisture and temperature, since there was only 72 mm of precipitation, and this was during the winter months when temperatures were low (Table 3). This agrees with the findings of Ou et al. (1982), who showed that carbofuran decomposition decreased as soil moisture and temperature decreased.

A universal question that should be asked before any substance is released into the environment is: "How long will it be detectable?" To estimate the time required (x in days) for the total amount of tebuthiuron in

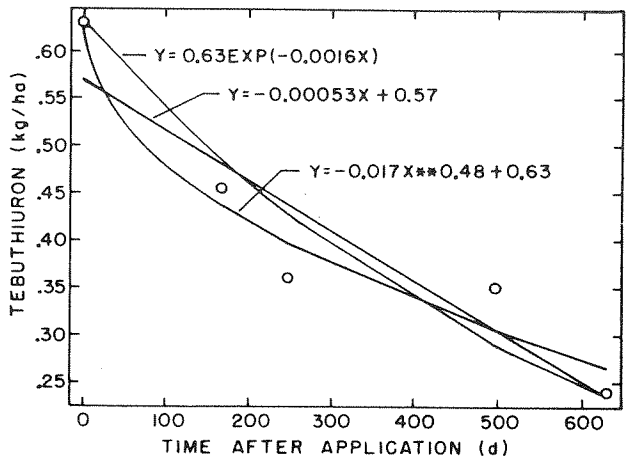

Fig. 2—Total tebuthiuron in the test plots with time and linear and nonlinear regression equations of the data.

the test plots (y in kg/ha) to reach 0.01 kg/ha, the detection limit in the test plots, the following linear and nonlinear regression equations were used:

$$y = mx + b, \quad [1]$$

where $m = -0.00053$ and $b = 0.57$, and

$$y = ax^b + c, \quad [2]$$

where $a = -0.017$, $b = 0.48$, and $c = 0.63$, and

$$y = 0.63e^{bx}, \quad [3]$$

where $b = -0.0016$.

A plot of the average total tebuthiuron in the two test plots vs. time, along with regression equations, is given in Fig. 2. The standard error for Eq. [1] was 0.064 kg/ha, and to reach 0.01 kg/ha in the test plots, it would take 2.9 yr by this equation. For Eq. [2], the standard error was 0.033 kg/ha, and using this equation, it would take 4.9 yr to reach a concentration of 0.01 kg/ha. Equation [3] predicted 7.2 yr to reach a concentration of 0.01 kg/ha, with a standard error of 0.045 kg/ha. The use of regression equations and extrapolating them is plagued with many problems, but it was felt that at least the linear and nonlinear regression equations would give a range of time for the tebuthiuron to disappear in the test plots. The large differences in the estimated time to reach the tebuthiuron detection limit in the test plots points out the difficulty in predict-

Table 3—Actual and average† precipitation on the study area between sampling dates of the test plots.

Date	Actual precipitation	Average precipitation
	mm	
3 Feb. 1981-19 July 1981	143	90
20 July 1981-07 Oct. 1981	183	149
08 Oct. 1981-15 June 1982	72	112
16 June 1982-26 Oct. 1982	188	222
Totals	586	573

† Eleven-year average, 1970-1980.

Table 4—Vegetative defoliation and apparent kill on the watershed after 217 and 615 d from an application of tebuthiuron at a rate of 0.84 kg/ha.†

Species	Defoliation		Kill
	217 d	615 d	
	%		
Tarbush (*Flourensia cernua*)	91	100	100
Creosote-bush (*Larrea tridentata*)	83	90	61
Whitethorn (*Acacia constricta*)	89	94	92
Graythorn (*Condalia spathulata*)	52	88	0
Fourwing saltbush (*Atriplex canescens*)	92	100	100
Littleleaf sumac (*Rhus microphylla*)	80	100	100

† Forage grass population too sparse to evaluate.

ing precisely the disappearance of chemicals released in the environment and the need for longer periods of record.

At least 52% of each brush species on the watershed was defoliated after 217 d (Table 4). Evaluation at 615 d showed a higher defoliation rate for all species, with 100% mortality of tarbush, fourwing saltbush, and littleleaf sumac. Although the graythorn was not killed, an estimated defoliation of 88% was obtained. Creosote-bush had 90% defoliation and 61% of the plants were killed. The percentage kill of the graythorn and creosote-bush should increase, since tebuthiuron residues remained in the soil, as indicated by the test plot analysis (Table 2). Jacoby et al. (1982) reported 98% kill of creosote-bush after 20 months with an application rate of 1.0 kg/ha.

SUMMARY AND CONCLUSION

The total tebuthiuron removed from the rangeland watershed in the runoff water and suspended sediment over two seasons was 0.47% of the amount applied (0.84 kg/ha). Tebuthiuron removal associated with the bedload sediment was negligible. The low total removal of tebuthiuron in runoff water was believed to be due to the timing of the application. Tebuthiuron was applied at a time when precipitation rates are expected to be less than the infiltration rate, but when winter precipitation would be sufficient to leach at least some of the chemical into the soil profile. Based on the result of this study, it would be recommended that this procedure be followed for tebuthiuron application to minimize the probability of removals in runoff.

Tebuthiuron moved to a depth of 15 cm in 8 months with 326 mm of precipitation, and remained at that depth despite additional precipitation. The total tebuthiuron remaining in the test plots after 21 months was 38% of that applied. Regression equations were developed that predicted the tebuthiuron would persist from 2.9 to 7.2 yr before the total amount in the test plots would fall below the detection limit. Significant differences in the estimated time required to reach the detection limit of tebuthiuron in the test plots emphasizes the difficulty in predicting precisely the disappearance of chemicals released in the environment and the need for longer periods of actual sampling and analysis.

Tebuthiuron, applied at a rate of 0.84 kg/ha, was effective in reducing the dominate brush species on the watershed, producing 100% kill for tarbush, fourwing saltbush, and littleleaf sumac after 615 d, and lower percentages for creosote-bush, whitethorn, and graythorn.

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