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Z. Liu
South Dakota State University

S. A. Clay
South Dakota State University, sharon.clay@sdstate.edu

J. Gaffney
South Dakota State University

D. Malo
South Dakota State University

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ATRAZINE AND ALACHLOR ADSORPTION CHARACTERISTICS TO BENCHMARK SOIL SERIES IN EASTERN SOUTH DAKOTA

Z. Liu, S.A. Clay, J. Gaffney, and D. Malo
Plant Science Department
South Dakota State University, Brookings, SD 57007

Abstract

Corn, grain sorghum, and soybean are grown on about six million acres in eastern South Dakota each year. Two herbicides used routinely for weed control are atrazine(6-chloro-*N*-ethyl-*N'*-(1-methylethyl)-1,3,5-triazine-2,4-diamine) in corn and grain sorghum and alachlor (2-chloro-*N*-(2,6-diethylphenyl)-*N*-(methoxymethyl)acetamide) in all three crops. Six benchmark soil series that include a majority of the cropped acres treated with these herbicides are the Egan, Moody, Nora, and Brandt silty clay loams, and Clarno and Enet loams. Batch adsorption studies determined atrazine and alachlor binding characteristics to these soils and aids in assessing the amount of herbicide available for movement. These data also provide a basis for future use and management decisions for these and other related herbicides in similar soils. Soils from three horizons (A, B, and C) for each soil type were treated with atrazine or alachlor at four herbicide concentrations. Atrazine and alachlor sorption partition coefficients differed most in A horizon soils and ranged from 2.16 to 5.35 $\mu mol^{1-1/n}$ $L^{1/n}Kg^{-1}$ for atrazine and 1.95 to 5.78 $\mu mol^{1-1/n}$ $L^{1/n}Kg^{-1}$ for alachlor. Atrazine binding to A horizon soils ranked as Brandt >Egan = Moody > Enet = Clarno > Nora. Alachlor binding to A horizon soils ranked as Brandt >Moody > Nora > Enet > Clarno. B and C horizon soils had lower binding for both herbicides; the sorption partition coefficient for atrazine ranged from 0.12 to 1.9 $\mu mol^{1-1/n}$ $L^{1/n}Kg^{-1}$ while alachlor ranged from 0.43 to 1.64 $\mu mol^{1-1/n}$ $L^{1/n}Kg^{-1}$.

These data indicate that some soil types would be more susceptible to herbicide leaching than others. Once the herbicide moves through the A horizon, it may move rapidly through the lower soil profile (because of the decrease in binding capacity), and therefore, increase the vulnerability of the aquifer to contamination. Best management practices for these herbicides are being investigated to limit their movement through soil.

INTRODUCTION

Since the late 1940's, agricultural production in the United States has depended on the use of chemicals (e.g. herbicide and insecticides) to control weeds and other pests. From 1992 to 1994, about 97 % of corn and soybean acreage in the U.S. were treated with one or more herbicide applications (Agricultural Statistics, 1995-1996). Short term benefits of herbicide application are weed control and increased crop yields. However, long term effects of herbicides on the environment have largely been ignored in crop production.

Corn, grain sorghum, and soybeans are grown on about 6 million acres in eastern South Dakota each year. Atrazine (6-chloro-*N*-ethyl-*N'*-(1-methylethyl)-1,3,5-triazine-2,4-diamine) and alachlor (2-chloro-*N*-(2,6-diethylphenyl)-*N*-(methoxymethyl)acetamide) are two herbicides which are relatively inexpensive and used routinely for weed control for corn, grain sorghum, or soybean in eastern South Dakota. In a survey of 16 eastern South Dakota counties conducted in 1990 and 1991, about 510,000 acres of corn were treated with atrazine, and 630,000 acres of corn and soybean were treated with alachlor (Table 1).

Undesirable environmental side effects such as detectable concentrations of herbicides in groundwater have become major national social and scientific concern. For example, US Environmental Protection Agency set primary drinking water standards at maximum contaminant levels of 3 ppb for atrazine and 2 ppb for alachlor (Code of Federal Regulations, 1993). In 1990, the National Survey of Pesticides in Drinking Water Wells found that about 10.4% of the community water supply wells and 4.2% of the rural domestic wells contain pesticides or pesticide degradates at or above the minimum reported limit used in the survey (Cohen, 1992). Both atrazine and alachlor have been detected in well water surveys conducted in Iowa, Minnesota, Nebraska and South Dakota (Hallberg, 1989).

Factors influencing herbicide detection in groundwater include point source problems, such as spills and back siphoning into wells, and nonpoint source contributions. Nonpoint source contamination may be from herbicides applied at normal use rates with low level herbicide movement through soil profiles and into aquifers. Herbicide movement into aquifers may be influenced by: 1) herbicide physical and chemical properties; 2) soil physical and chemical properties such as soil texture, soil permeability, soil hydraulic properties, depth to the aquifer, soil pH and organic carbon content; 3) environmental parameters such as rainfall; and, 4) crop management such as tillage.

Herbicide movement through soil is affected by its binding capacity to soil. Soil properties (e.g., organic matter, pH, and clay content) have a significant effect on herbicide adsorption to soil (Koskinen and Harper, 1990). In general, as herbicide binding capacity to soil increases, herbicide movement through the soil decreases. Kladivko et al. (1991) reported that the concentration of herbicides in subsurface drainflow corresponded to the rank-order of batch adsorption coefficients measured in the laboratory.

The objective of this study was to investigate atrazine and alachlor adsorption characteristics for benchmark soils in eastern South Dakota. The data obtained will aid in assessing the amount of herbicide available for movement and, ultimately, the vulnerability of groundwater to contamination by these herbicides. These data also provide a basis for future use and management decisions for these and other related herbicides in similar soils.

MATERIALS AND METHODS

Adsorption

Six benchmark soil series used in this study were collected from five eastern counties of South Dakota. Four silty clay loam (scl) and two loam (l) soils were chosen and included Egan scl (fine-silty, mixed, mesic Udic Haplustolls) and Nora scl (fine-silty, mixed, mesic Udic Haplustolls) from Lincoln county, Moody scl (fine-silty, mixed, mesic Udic Haplustolls) from Minnehaha county, Brandt scl (fine-silty, mixed, mesic Udic Haplustolls) from Brookings county, Clarno l (fine-loamy, mixed mesic Udic Haplustolls) from Hutchinson county and Enet l (fine-loamy over sandy or sandy-skeletal, mixed, Mesic Pachic Haplustolls) from Clay county. The soil was collected by horizon, air-dried, and passed through a 2-mm sieve. Herbicides were dissolved in methanol(< 1 mL) and then diluted to final concentration of 2.5, 5.0, 10.0, or 20.0 μmol L^{-1} of herbicide in 0.01 $\underline{M}$ $CaCl_2$. Each solution then was spiked with uniformly-ring-labeled-^{14}C-atrazine/alachlor in the range of 0.1 to 1.2 kilobecquerel (KBq). Ten g of soil were placed in a centrifuge tube and 5 mL of 0.01 $\underline{M}$ $CaCl_2$ solution and 5 mL of ^{14}C- herbicide solution were added.

After herbicide addition, a one-day batch equilibration study was conducted by: (i) mechanically shaking the treated soil slurry for 24 h at 25°C; (ii) centrifuging the slurry at 8,000 r.p.m (approx. 8,000 g) for 20 minutes to separate supernatant and soil; (iii) pipetting out the supernatant, and (iv) adding scintillation cocktail and counting ^{14}C in solution using a Packard 1600TR Liquid Scintillation Analyzer. The amount of herbicide adsorbed to soil was determined by the difference between the amount radioactivity in the initial solution and the radioactivity in the solution after soil equilibration. The linear form of the Freundlich equation was used to describe herbicide sorption to soil (Koskinen and Harper, 1990) . The linear form of the Freundlich equation is :

$$\log [C_s] = \text{Log Kf} + 1/n \ \text{Log} [C_e];$$

where, C_s is the μmol herbicide adsorbed to per kg soil;

C_e is the umol herbicide in per L of supernatant solution after equilibration;

Kf and 1/n are empirical constants.

Data Analysis

Freundlich sorption isotherm coefficients were calculated by the least squares technique using the log-transformed equilibrium data. Statistical evaluation included comparison of the slopes and intercepts of the regression lines, and calculation of the 95% confidence intervals for the intercept (log K_f) and slope (1/n).

RESULTS AND DISCUSSION

Atrazine and Alachlor usage. The acres of corn and soybean in 16 counties of eastern South Dakota treated with atrazine and alachlor in 1990 and 1991 are listed in Table 1. The percentage of corn fields treated with atrazine ranged from 5% to 75%. Lincoln (75%), Union (60%), Moody (50%) and Minnehaha (45%) counties had the highest percentages of corn treated with atrazine. The percentage of corn and soybean fields treated with alachlor ranged from 5% to 50%, with the highest percentage of use in McCook and Clay (50%), Hutchinson (40%), Lake and Spink (33%), and Moody (30%) counties. The combined atrazine and alachlor usage was over 1.1 million acres, and accounted for 42% of total corn and soybean acres. The survey also estimated that the usage of atrazine and alachlor from 1991 to 1996 would remain stable or even increase in Moody, Minnehaha, Lincoln, and Hutchinson counties.

Soil Characteristics. The six soil series from eastern South Dakota were chosen because they represent a large portion of the acreage treated with alachlor and/or atrazine (Table 2). The physical characteristics of these soils are reported Table 3. The soil pH in A horizon soil ranged from 5.50 to 7.21

Table 1. 1990–1991 survey results of atrazine and alachlor usage in 16 eastern South Dakota counties[1].

County	Acres of Corn	% of corn acres treated with Atrazine	Acres of Corn + Soybean	% of acres of corn and soybean treated with Atrazine
Lincoln	133,000	75	268,000	5
Union	107,000	60	195,000	20
Moody	118,000	50	118,000	30
Minnehaha	150,000	45	260,000	17.5
Spink	128,000	40	219,000	33
Lake	108,000	33	186,000	33
McCook	95,000	20	180,000	50
Hutchinson	151,000	15	260,000	40
Clay	84,000	7.5	170,000	50
Lyman	13,900	45	13900	17
Turner	130,000	20	262,000	20
Charles Mix	71,000	17	not determined	not determined
Brown	130,000	10	130,000	10
Yankton	228,000	5	228,000	5
Brookings	127,000	7.5	127,000	4.5
Tripp	33,000	15	36,000	0.5

[1] Surveyed by Jim Gaffney, former graduate research assistant, Personal Communication with S.A. Clay

Table 2. The percent coverage of the six benchmark soil series chosen for this study across eastern South Dakota.

Soil Association	**Counties Covered**	**% Coverage**
Clarno/Crossplain-/Bonilla-/Prosper-/ loam	McCook, Hutchinson	36.3
Egan/Wentworth-/Beadle-/silty clay loam	Lake, Lincoln, Moody	14.6
Moody/Nora-/silty clay loam	Minnehaha, Moody	11.1
Nora/Moody-/Crofton-/silty clay loam	Minnehaha, Union	12.8
Enet loam	Clay	<1
Brandt/Estilline silty clay loam	Brookings, Codington, Deuel, Hamlin, Moody, Minnehaha	5

Table 3. Selected soil properties for six benchmark soils in eastern South Dakota.

Soil Series	**Sampled County**	**Horiz**	**Depth (in)**	**pH (1:1)**	**OC %**	**Sand %**	**Silt %**	**Clay %**
Moody silty clay loam	Minnehaha	Ap	0-7	6.39	2.47	9.65	56.9	34.6
		Bw	7-17	7.17	0.55	21.0	54.6	24.6
		Bk	30-42	7.96	0.27	25.7	56.6	17.8
		Ck	42-56	8.20	0.22	7.45	75.1	17.5
Nora silty clay loam	Lincoln	Ap	0-7	7.21	2.02	9.2	62.0	28.9
		Bw	7-18	7.40	0.51	11.1	62.3	26.7
		Bk	18-30	7.76	0.24	10.5	65.3	25.3
Clarno loam	Hutchinson	Ap	0-9	6.56	1.51	30.4	40.6	29.1
		Bw	9-16	7.01	1.10	31.9	38.4	29.6
		Bk	16-36	7.84	0.54	36.7	38.7	24.6
		C	36-60	7.96	0.05	37.0	39.0	24.2
Egan silty clay loam	Lincoln	Ap	0-8	5.81	3.25	7.55	55.9	36.6
		Bw	8-25	6.61	2.22	4.25	61.2	34.6
		Bk(2C)	25-30	7.93	0.84	9.9	53.4	36.8
		2C(Bk)	30-60	7.91	0.22	30.3	37.3	32.5
Enet loam	Clay	Ap	0 -6	6.6	2.6	27.5	46.5	26.0
		AB	6 - 12	6.0	1.7	30.4	41.4	28.2
		B	12 -18	6.3	1.17	23.2	49.4	27.4
Brandt silty clay loam	Brookings	A	0 - 16	5.5	4.5	16.2	54.4	29.4
		B	16 -51	6.2	1.0	11.8	56.5	31.7
		C	51+	7.9	0.2	17.2	52.9	29.5

and ranked as Nora scl>Enet l = Clarno l = Moody scl > Egan scl > Brandt scl. The soil pH ranged from 6.61 to 7.96 in B horizon soil and 6.20 to 8.25 in C horizon soil. The % carbon (%C) in A horizon soil ranged from 1.53 to 4.50 and ranked as Brandt scl > Egan scl > Enet l = Moody scl > Nora scl > Clarno l. The %C ranged from 0.24 to 2.84 in B horizon soil and 1.50 to 3.31 in C horizon soil. The % sand content in A horizon ranged from 7.55 to 30.4 and ranked as Clarno l > Enet l > Brandt scl > Moody scl = Nora scl > Egan scl.

Atrazine sorption. Atrazine sorption characteristics for the six soils chosen for this study are reported in Table 4 and Figures 1 & 2. The Kf value is the amount of herbicide adsorbed to the soil when the amount in solution is 1 µmol L^{-1}. It is an index of herbicide binding capacity to soil with lower Kf values indicating less herbicide sorbed to soil. The atrazine Kf values differed most in A horizon soils and ranged from 2.16 to 5.35. Atrazine bind-

Table 4. Atrazine Adsorption Isotherms for six benchmark soils in eastern South Dakota.

Soil type	**Sampled County**	**Horiz**	**Depth (in)**	**Kf***	**l/n****	**r^2**
Moody silty clay loam	Minnehaha	Ap	0 - 7	3.72 (3.47 -3.99)	0.86 (0.03)	0.99
		Bw	7 - 17	0.92 (0.89 -0.94)	0.94 (0.01)	0.99
		Bk	30 - 42	0.63 (0.56 -0.70)	0.88 (0.05)	0.98
		Ck	42 - 56	0.56 (0.53 -0.60)	0.96 (0.03)	0.99
Nora silty clay loam	Lincoln	Ap	0 - 7	2.16 (2.06 - 2.26)	0.90 (0.02)	0.99
		Bw	7 - 18	1.02 (0.996 - 1.06)	0.89 (0.01)	0.99
		Bk	18 - 30	0.65 (0.60 -0.69)	0.94 (0.03)	0.99
Clarno loam	Hutchinson	Ap	0 - 9	2.80 (2.62 - 2.98)	0.85 (0.03)	0.99
		Bw	9 - 16	1.57 (1.51 -1.63)	0.91 (0.02)	0.99
		C	36 - 60	0.61 (0.56 -0.65)	0.91 (0.03)	0.99
Egan silty clay loam	Lincoln	Ap	0 - 8	4.13 (3.68 -4.64)	0.89 (0.05)	0.98
		Bw	8 - 25	1.91 (1.78 -2.05)	0.88 (0.03)	0.99
		Bk	25 - 30	0.95 (0.88 -1.04)	0.93 (0.04)	0.99
Enet loam	Clay	Ap	0 - 6	2.93 (2.81 - 3.06)	0.80 (0.01)	0.99
		AB	6 - 12	2.29 (2.12 -2.47)	0.80 (0.01)	0.99
		Bw	12 -18	0.81 (0.73 -0.90)	0.84 (0.01)	0.99
Brandt silty clay loam	Brookings	A	0 - 16	5.35 (5.16 -5.55)	0.82 (0.01)	0.99
		B	16 - 51	1.06 (0.98 - 1.14)	0.87 (0.01)	0.99
		C1	51 +	0.12 (0.08 - 0.18)	0.93 (0.05)	0.97

* Numbers in parentheses are the 95% confidence interval (CI) for Kf.

** Numbers in parentheses are the standard error of 1/n.

ing to A horizon soils ranked as Brandt scl > Egan scl = Moody scl > Enet l = Clarno l > Nora scl. These data indicate that about twice as much atrazine is in solution of the Nora scl and Clarno l than Egan and Brandt scl.

The lower atrazine sorption to the Nora scl and Clarno l may be due to the higher soil pH and lower organic carbon than Brandt scl. A negative correlation between Kf and pH was calculated for A horizon soils (r = -0.96; $p<0.05$) (Fig. 3 and Table 5). These data indicate that as soil pH increases atrazine sorption decreases. Soil pH has been shown to affect atrazine binding to soil with more sorption to high than low pH soils (Clay et al., 1988; Liu et al.,1995). This is due to the fact that atrazine is slightly basic and at lower pH, the molecule has slightly positive charge, resulting in greater sorption. A positive correlation of Kf with organic carbon content (r = 0.91 $p<0.05$) was observed (Fig. 4 and Table 5) so that as %C increases so did atrazine sorption. The high sand content of the Clarno l also may have contributed to lower atrazine sorption than in other soils.

Atrazine binding to soil decreased as the soil

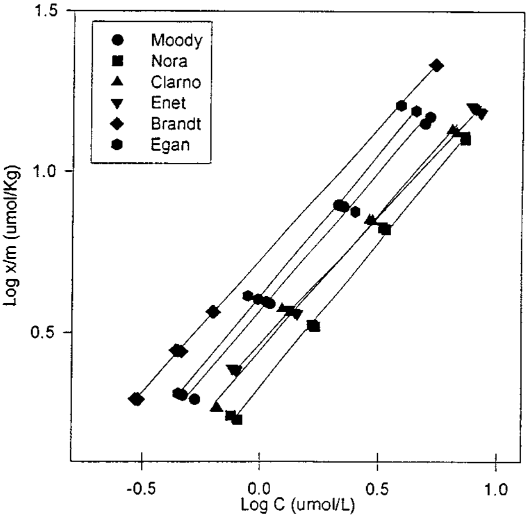

Figure 1. Atrazine sorption in A horizon soils.

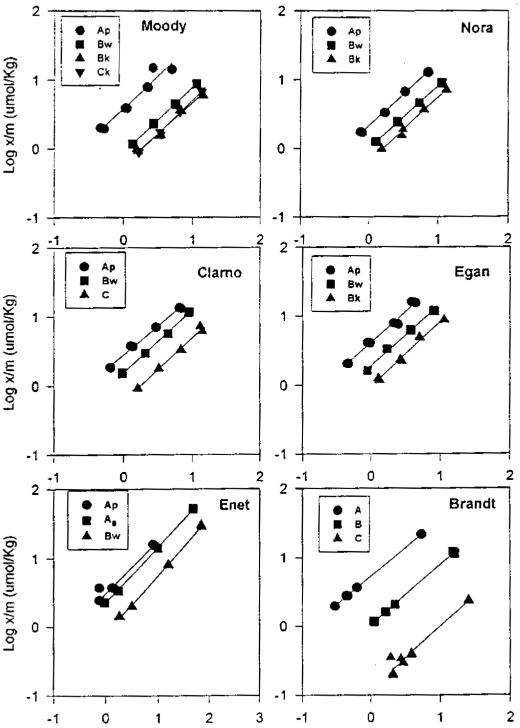

Figure 2. Atrazine sorption in six benchmark soils.

depth increased most likely due to the increase in soil pH and sand content. The atrazine Kf value ranged from 0.63 to 1.91 in B horizon soils and 0.12 to 0.61 in C horizon soils. The lower Kf values in B and C horizons indicate that once the herbicide moves through the A horizon soil, it is less sorbed, and perhaps more readily transported through the lower soil horizons.

The 1/n value for all six soil series were less than 1 and ranged from 0.79 to 0.94. No significant differences in 1/n values were noted among soils or soil horizons studied. The 1/n value is the slope of the herbicide sorption isotherm. When 1/n is less than 1, this indicates that the amount of herbicide sorbed to soil is dependent of initial herbicide concentration in solution with less herbicide sorbed to soil as the initial concentration increases. However, when 1/n = 1, the percent of herbicide sorbed to soil is independent of initial concentration in solution.

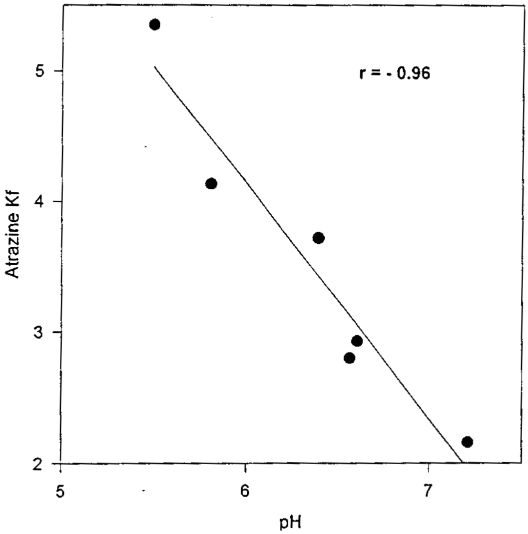

Figure 3. Correlation of atrazine Kf with soil pH.

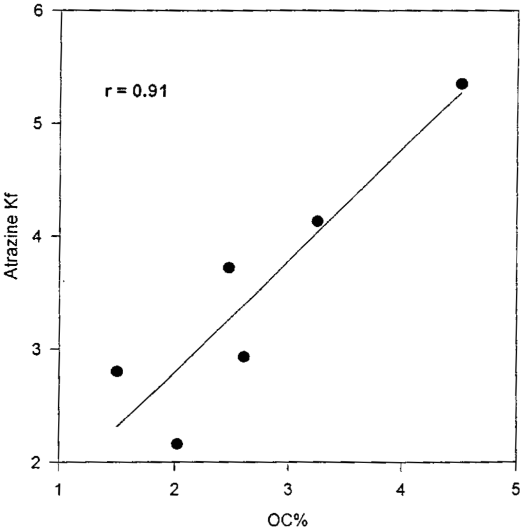

Figure 4. Correlation of atrazine Kf with soil OC%.

Table 5. Correlation of Kf values with pH and organic carbon content for A horizon soils.

	Kf atrazine **r**	**Kf Alachlor** **r**
pH	-0.96	-0.86
% Organic Carbon	0.91	0.93

Alachlor sorption: Alachlor sorption characteristics for five soil series (Egan scl not included) in eastern South Dakota are reported in Table 6 and Figures 5 & 6. The alachlor Kf values differed most in A horizon soils and ranged from 1.95 to 5.78. Alachlor binding to A horizon soils ranked as Brandt scl > Moody scl >Nora scl > Enet l > Clarno l. These data indicate that about three times as much alachlor is in solution in the Clarno l than Brandt scl. A positive correlation of Kf with organic carbon content for A horizon soils was calculated ($r = 0.93$, $p<0.05$) (Table 5 & Fig. 7) and a negative correlation of Kf with pH was observed ($r=-0.86$, $p=0.09$) (Table 5). The positive correlation with organic carbon agrees with Peter and Weber (1985) who reported that soil organic matter content was highly correlated with sorption of alachlor. Alachlor sorption was not as strongly affected by pH as atrazine because alachlor is a neutral molecule.

Alachlor binding to soil decreased rapidly as the soil depth increased. The alachlor Kf value ranged from 1.06 to 1.64 in B horizon soils and 0.43 to 0.77 in C horizon soils. The lower Kf values in B and C horizons indicate that less alachlor was sorbed to B and C horizon soils and if alachlor moved

Table 6. Alachlor Adsorption Isotherms for six benchmark soils in eastern South Dakota.

Soil type	**Sampled County**	**Horiz**	**Depth (in)**	**Kf ***	**1/n****	**r2**
Moody silty clay loam	Minnehaha	Ap	0 - 7	3.36 (3.04 -3.70)	0.72 (0.01)	0.99
		Bw	7 - 17	1.63 (1.19 -2.24)	0.64 (0.05)	0.97
		Bk	30 - 42	1.14 (0.78 -1.67)	0.71 (0.06)	0.96
Nora silty clay loam	Lincoln	Ap	0 - 7	2.72 (2.53 - 2.94)	0.76 (0.01)	0.99
		Bw	7 - 18	1.55 (1.43 - 1.68)	0.73 (0.01)	0.93
		Bk	18 - 30	1.18 (0.70 -2.00)	0.85 (0.09)	0.99
Clarno loam	Hutchinson	Ap	0 - 9	1.95 (1.68 - 2.27)	1.06 (0.07)	0.97
		Bw	9 - 16	1.64 (1.50 -1.79)	0.91 (0.04)	0.99
		C	36 - 60	0.77 (0.73 - 0.80)	0.98 (0.03)	0.99
Enet loam	Clay	Ap	0 - 6	2.39 (2.29 - 2.50)	0.75 (0.01)	0.99
		AB	6 - 12	2.10 (1.86 -2.37)	0.73 (0.02)	0.99
		Bw	12 -18	1.06 (0.86 -1.30)	0.76 (0.03)	0.98
Brandt silty clay loam	Brookings	A	0 - 16	5.78 (4.94 -6.75)	0.72 (0.01)	0.99
		B	16 - 51	1.88 (1.69 - 2.10)	0.86 (0.01)	0.99
		C1	51 +	0.43 (0.25 - 0.71)	0.72 (0.12)	0.90

* Numbers in parentheses are the 95% confidence interval (CI) for Kf.

** Numbers in parentheses are the standard error of 1/n.

out of the A horizon, it has the potential to move rapidly through the lower soil profile.

The 1/n value for four of the five soil series were < 1. The exception was that Clarno l had an 1/n of 1.06, which indicated that the percent of alachlor adsorbed to soil is independent of initial alachlor concentration in solution.

CONCLUSIONS

The A horizon of the Enet l and Clarno l were the two soil types that had the lowest binding capacity for atrazine and alachlor while Brandt scl had the highest binding capacity for both atrazine and alachlor. These data indicate that the loam soils may be more susceptible to atrazine and alachlor movement through the A horizons than silty clay loam soil. Ranking of the soils for sorption of both herbicides was similar except the Nora scl that had less binding of atrazine than alachlor. The low sorption capacity of the Nora scl for atrazine is most likely due to the relatively high soil pH compared to the other soils.

Due to the decrease of atrazine and alachlor binding capacity to B and C horizon soils, once the herbicides move through the A horizon, they have

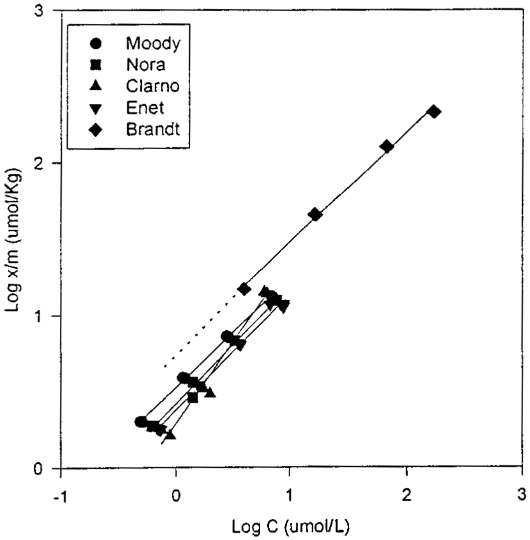

Figure 5. Alachlor sorption in A horizon soils.

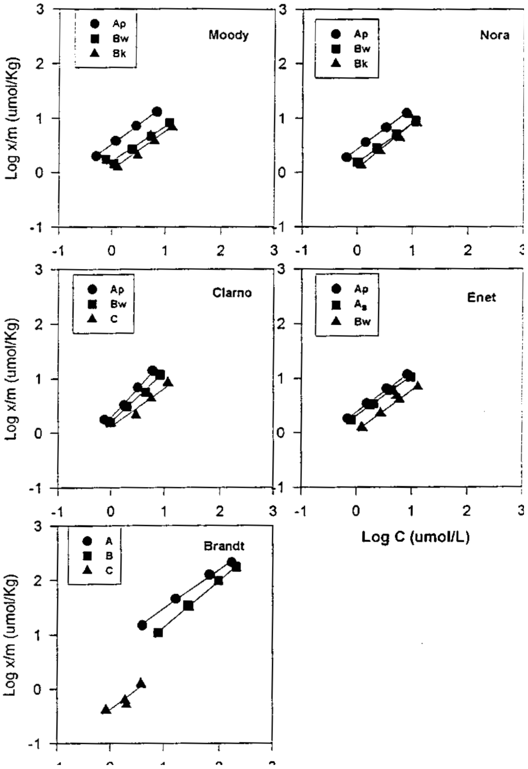

Figure 6. Alachlor sorption in five benchmark soils.

the potential to move rapidly through the B and C horizons. Herbicide degradation is slower (2 to 1000 times slower) as depth in the soil increases (Clay et al., 1997; Yen et al., 1994; Pothuluri et al., 1990). The low sorption and slow degradation properties increase the potential of these herbicides to contaminate shallow aquifers. Therefore, best management practices for these herbicides must be investigated to limit their movement through soil.

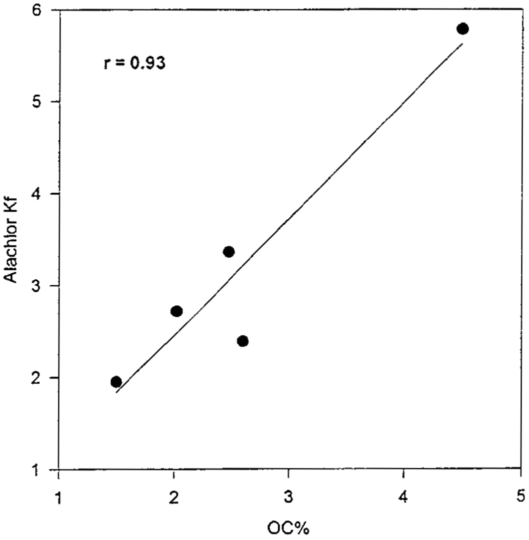

Figure 7. Correlation of alachlor with soil OC%.

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