Reprinted from the *Journal of Environmental Quality*
Volume 26, no. 5, Sept.–Oct. 1997, Copyright © 1997, ASA, CSSA, SSSA
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Atrazine Sorption at the Field Scale in Relation to Soils and Landscape Position

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ABSTRACT

Understanding the spatial variation of herbicide sorption in soils is important in determining the potential for leaching at the field scale. Our objectives were to determine the spatial variability of atrazine sorption (6-chloro-*N*-ethyl-*N*′-(1-methylethyl)-1,3,5-triazine-2,4-diamine) at the field scale and to relate sorption partition coefficients (K_d) to landscape position and soil survey map units. Atrazine sorption was measured on 241 surface samples from a 6.25-ha field using batch-equilibration methods. Field-scale variability in atrazine sorption coefficients was described using spherical semivariograms. Less than 20% of the total semivariance in atrazine K_d values was found at lag distances <10 m, indicating there was relatively little variability at this scale. Multiple regression analyses using pooled data revealed that atrazine sorption was influenced by soil organic C, pH, and, to a lesser extent, soil clay. We also evaluated the relationship of atrazine sorption to landscape position and soil series. Less atrazine was sorbed by soils from upland shoulder slopes than by soil in level and depressional areas (potholes). Soils from foot slope and back slope landscape positions were intermediate in atrazine sorption. The magnitude of atrazine sorption by soils in different landscape positions was also related to variations in soil organic C content, pH, and clay content. The greater sorption of atrazine by soils in the pothole depressions may reduce transport of this herbicide to tile lines that commonly drain these soils. Field-scale maps of atrazine sorption based on landscape position more effectively predicted the actual distribution of the K_d values determined by kriging, than did maps based on soil series.

ATRAZINE is commonly used in corn (*Zea mays* L.) production in the Midwest. In 1990, 61% (roughly 3 million ha) of the corn in Iowa was treated, corresponding to an application of approximately 3.4 million kg of atrazine (Hartzler and Wintersteen, 1991). Atrazine is present year round in groundwater and streams in Iowa and other midwestern states (Thurman et al., 1991; Burkart and Kolpin, 1993). Atrazine enters surface water via runoff (Hall, 1974; Hall et al., 1991) and subsurface tile drainage (Jayachandran et al., 1994). In addition, shallow groundwater may be contaminated with atrazine during exchanges with surface water (Squillace and Thurman, 1992).

Identification of soils that are more vulnerable to herbicide leaching is a key step in designing management practices that reduce groundwater pollution. This will require knowledge of the spatial distribution of soil properties and processes that control herbicide movement. Sorption of herbicides to soil is one of several processes that control herbicide losses in leaching and runoff. Earlier studies show that herbicide sorption (Rao et al., 1986; Wood et al., 1987) and persistence (Sadeghi and Isensee, 1992) vary substantially at the field scale. Watershed-scale assessments of soil vulnerability to pesticide leaching have used information on soil properties based on soil map units in combination with models that simulate herbicide movement (Khan and Liang, 1989; Loague et al., 1990). While the impact of uncertainty in soil properties within higher-order taxonomic units has been recognized (Loague et al., 1990), less attention has been paid to variability within soil series at the field scale or to the position of soil within the landscape.

The improvement of computerized mapping techniques, including Geographical Information Systems (GIS) technology, now allows the use of county-level soil survey maps as base maps for vulnerability assessments and for precision application of pesticides and nutrients. However, soil map units are recognized to be heterogeneous at many scales (Edmonds and Lentner, 1986; Seyfreid et al., 1992). Alternatively, landscape position has been used to classify soils, and the linkage between landscape position and the distribution of soil map units has long been recognized (Walker and Ruhe, 1968). Landscape position analysis in combination with soil survey maps has been used to develop high resolution spatial estimates of soil properties (Moore et al., 1993.) However, the use of soil survey maps or landscape position classes to evaluate herbicide sorption at the field scale has not been evaluated.

Our study was undertaken to (i) describe the spatial variability of atrazine sorption at the field scale and (ii) evaluate landscape position classes and soil map unit delineations for their potential use as predictors of atrazine sorption at the field scale.

MATERIALS AND METHODS

Site Description and Soil Sampling

The study was conducted in a 6.25-ha field located in the western part of the Walnut Creek watershed, Boone County, Iowa. The landscape features and soils are characteristic of the Des Moines lobe and contain examples of closed depressions surrounded by nearly level to gently sloping upland areas that are typical of central Iowa (Walker and Ruhe, 1968). The soils consist mainly of well- to poorly-drained Mollisols. The family taxonomic classification for the Mollisols mapped in the study area is shown in Table 1. Slopes in the field range from <1% in the depressions to >3% in the uplands. The field site is distinguished by two closed depressions, one of which is drained by a subsurface tile-drain (Cambardella et al., 1994). In our study, 121 surface (0–15 cm) soil samples were collected at 25-m intervals from a 250 by 250 m square grid (6.25 ha) during April of 1992 (Fig. 1A). In addition, 120 samples were collected at distances of 2, 5, and 10 m south and east from 20 of the main grid nodes (Fig. 1A), bringing the total sample number to 241. Each sample was a composite consisting of three cores collected within a 1-m radius circle surrounding each point. Detailed descriptions of the site; sampling procedures; and soil organic C, pH, and particle-size analyses of

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Published in J. Environ. Qual. 26:1271–1277 (1997).

Abbreviations: K_d and K_{oc}, sorption partition coefficients; HPLC, high performance liquid chromatography.

Table 1. Family taxonomic classification of soil series mapped in the study area.

Soil series	Family taxonomic classification
Canisteo	Fine-loamy, mixed (calcareous), mesic Typic Haplaquoll
Clarion	Fine-loamy, mixed, mesic Typic Hapludoll
Harps	Fine-loamy, mesic Typic Calciaquoll
Nicollet	Fine-loamy, mixed, mesic Aquic Hapludoll
Okoboji	Fine, montmorillonitic, mesic Cumulic Haplaquoll

the soil samples have been previously reported (Cambardella et al., 1994).

Landscape Position and Map Unit Assignments

The landscape position assignments (Fig. 1B) for each of the 121 main grid (elevation measurements at 25-m increments in the 250 by 250 m grid) points were determined using algorithms developed by Pennock et al. (1987) with the following modifications. Positions with concave profiles (curvature < -0.04°/m) were classified as foot slopes and those with convex profiles (curvature > 0.04°/m) were classified as shoulder slopes. Profile curvatures between −0.04 and 0.04°/m were classified as linear. Linear landscape elements were further subdivided by slope gradient. Gradients <0.60° were classified as level. Level landscape elements within the study area were further subdivided into level upland areas and level areas found in the closed drainage depressions (potholes). Positions with gradients >0.60° were classified as back slopes. The degrees of profile curvature and gradient, which differentiate the landscape positions, are less than those used by Pennock et al. (1987) in Saskatchewan because the central Iowa landscape has less relief and gentler slopes than the Saskatchewan landscape.

Map unit assignments were made by superimposing a digitized, county-level, soil survey map (Andrews and Dideriksen, 1981) of our field over the sampling grid to identify the location of each sampling point with respect to the map unit boundaries (Fig. 1A). The positioning of soil map units relative to the sampling grid was confirmed using distances from the edges of the grid to adjacent roads for reference.

Sorption and Soil Properties

Atrazine K_d values were determined by batch equilibration on duplicate 4-g samples of 2-mm sieved air-dried soil for the 241 soil samples. Soil was equilibrated for 72 h with 15 mL solution containing 1.5 mg L^{-1} atrazine dissolved in 0.01 *M* $CaCl_2$. Preliminary experiments (data not shown) indicated that near-equilibrium conditions were established in 72 h. Atrazine concentrations were analyzed using HPLC as outlined by Novak et al. (1994). No atrazine metabolites were observed during the equilibration, and sorption to glassware was negligible. Sample means were calculated from duplicate analyses of atrazine K_d, and the sorption determination was repeated if the coefficient of variation (CV) exceeded 20%. The atrazine K_d (L kg^{-1}) sorption coefficient was calculated as K_d = mg of atrazine kg^{-1} of soil/mg of atrazine L^{-1} of equilibrium solution. Atrazine K_{oc} values were calculated by normalizing the K_d values for organic C ($K_{oc} = [K_d / \% \text{ organic C}] \times 100$).

Statistical Methods

The 241 measurements of atrazine sorption (K_d and K_{oc}), organic C, pH, and clay content were tested for normality using the procedures of D'Agostino et al. (1990) and found to be non-normal (Cambardella et al., 1994). Data were log-transformed prior to subsequent analyses to minimize the effects of the extreme values found in the log-normal distribution. This transformation resulted in normal distributions for K_d and K_{oc}. We used procedures described by Cressie (1993) and Hamlett et al. (1986) to check for nonstationarity in K_d and K_{oc}. Semivariograms for atrazine K_d and K_{oc} were estimated by least-squares regression (GS+, Geostatistics for the Agronomic and Biological Science, Version 1.1, Gamma Design

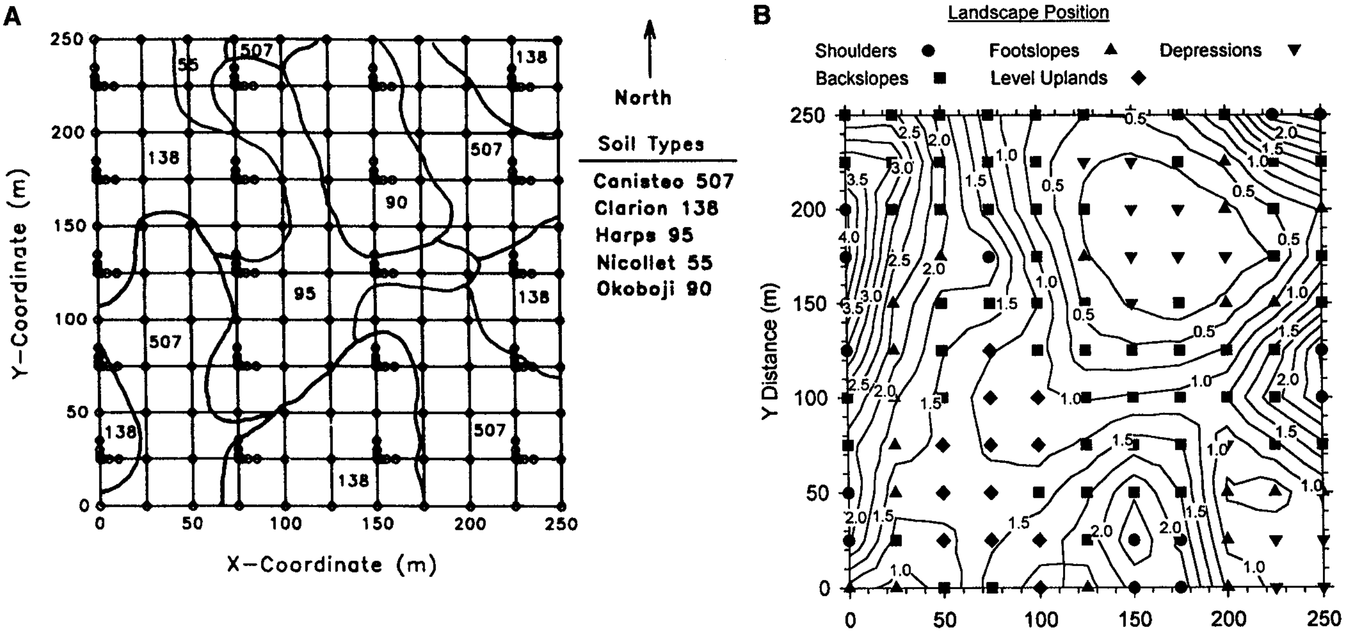

Fig. 1. (*A*) Map showing the sampling grid (25 by 25 m) in relation to soil series position (sampling locations shown as dots). (*B*) Landscape position assignments at points on a 25 by 25 m grid spacing. Contours indicate elevation (m) relative to the lowest point in the field. The highest point in the sampling grid was located on the western edge (0, 175). The large depressional area is centered at approximate coordinates of 175, 175 and a smaller one at 250, 0.

Software, St. Plainwell, MI 49080[1]) using spherical models, which were selected on the basis of model r^2 and residual sums of squares. Directional semivariograms (0, 45, 90, and 135°) were constructed to check for anisotropy. Omnidirectional semivariance calculations were based on data with a maximum lag distance of 125 m, divided into 31 lag classes, separated by an average of 4 m. Each lag class contained at least 40 pairs of points for calculation of semivariances, and most lag distance classes contained over 100 pairs of points.

Field distributions of atrazine K_d and K_{oc} (and organic C, pH, and clay content) were estimated by kriging, using either 5 by 5 m or 25 by 25 m blocks. The smaller block size allows construction of a fine-scale map that shows the variability of the sorption parameters. The larger sized blocks were used to be consistent with the landscape position classifications performed using elevation data from the center of each block. In order to estimate atrazine K_d and K_{oc} values for soils in each of the different landscape positions, we calculated pooled means from the kriged estimates for the 25 by 25 m blocks within each landscape position. In the calculation of the pooled mean and variance, block estimates were weighted by the inverse of their kriged variances. The variances for each block mean include both the random and spatial variance components. This technique for calculation of the pooled means and variances reduces the effects of sampling density and sample position and minimizes spatial dependence. We assume that the block means behave as independent estimators of the measured parameters. Similar data analyses were performed to estimate these parameters for each soil series mapped in the field.

RESULTS AND DISCUSSION

Spatial Variability of Atrazine Sorption

Semivariograms for K_d and K_{oc} show strong spatial dependence with low nugget variances and fairly large range values (Fig. 2). Preliminary tests indicated that the K_d and K_{oc} data met the assumptions of stationarity. Row (east-west) and column (north-south) means of K_d and K_{oc} (log-transformed data) showed no trend with distance, and row and column variances were generally independent of the magnitude of the mean (Hamlett et al., 1986). Directional semivariograms for all variables (not shown) showed only minor differences in sills and ranges, indicating that the semivariances were isotropic. Nugget values represent measurement error and the spatial variability present at lag distances smaller than the 2.6-m distance in the first lag class of the semivariogram. The low nugget value and high sill-to-nugget ratio indicate that there is little small-scale variability in atrazine K_d. Less than 21% of the total semivariance in K_d is present at lag distances <10 m. The nugget value for K_{oc} contributed 27% of the total semivariance of K_{oc}. Range values indicate the greatest distance over which parameters are spatially correlated, and these are similar in magnitude to ranges reported for metolachlor sorption to soil at a different site (Wood et al., 1987).

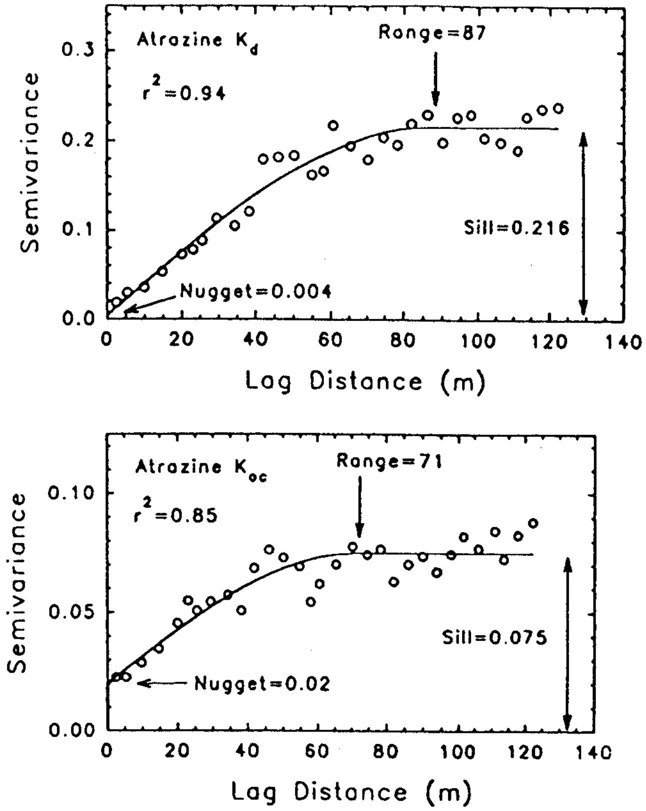

Fig. 2. Semivariances for the log-transformed parameters K_d and K_{oc} and the spherical model regressions (solid lines) describing the semivariograms for these parameters.

Atrazine K_d values, presented as contour maps based on 5 by 5 m block estimates, showed distinct spatial trends in atrazine sorption over the study area (Fig. 3). Increasing block size produces estimates averaged over larger areas, but general pattern of atrazine sorption in the field is the same as that obtained with smaller block sizes (Fig. 4). Kriged estimates of K_d for the 5 by 5 m blocks ranged from 1.7 to 14.4, and the block standard deviations ranged from 1.00 to 1.02. For the 25 by 25 m blocks, estimates of K_d ranged from 1.9 to 12.5, with

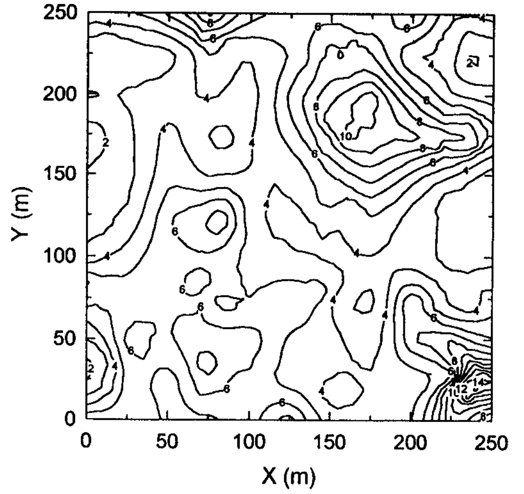

Fig. 3. Distribution of atrazine K_d represented as sorption isolines in a 6.25-ha field. The contour map was prepared from 2601 block-kriged estimates of atrazine K_d based on 5 by 5 m grid pattern using block estimates.

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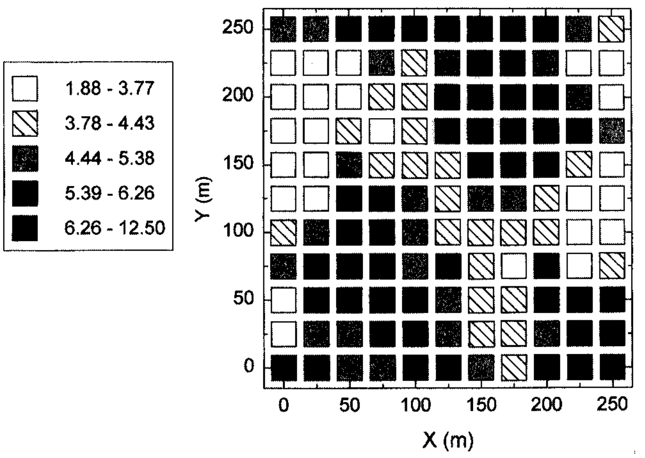

Fig. 4. Distribution of atrazine K_d based on 121 kriged estimates of atrazine K_d, with each square representing a 25 by 25 m block. The number of K_d values in each class represents 20% of the estimates.

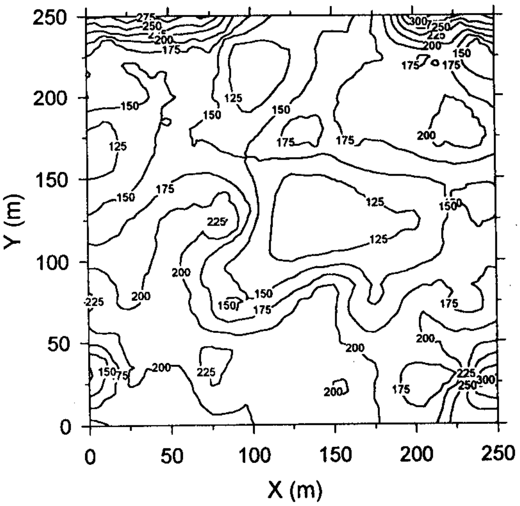

Fig. 5. Distribution of atrazine K_{oc} based on 2601 block-kriged estimates that are based on a 5 by 5 m block estimate.

the standard deviations ranging from 0.53 to 0.72. Increasing block size decreases the estimation error, but both block sizes have relatively small standard deviations due to the high density of sampling at the site.

Multivariate regression, using measured data from the 241 soil samples, showed that organic C, pH, and, to a lesser extent, clay content were able to predict atrazine sorption:

$$K_d = 2.01(\% \text{ organic C}) - 1.19 \text{ (pH)} + 0.04 \text{ (\% clay)} + 5.95, \quad r^2 = 0.76, P < 0.001.$$

Inclusion of pH and clay content raised the regression r^2 from 0.58 obtained with organic C alone to 0.76. Previous studies have identified the role of organic matter, pH, and clay in atrazine sorption (Dunigan and McIntosh, 1971; Laird et al., 1992). Variation in both the types and quantity of organic matter and clay can contribute to the strength of sorption.

The semivariogram of K_{oc} (Fig. 2) is essentially the spatial variability of atrazine sorption after detrending for organic C. The spatial distribution of K_{oc} (Fig. 5) differs from that of K_d (Fig. 3). Variation in K_{oc} represents the effects of soil constituents other than organic C that influence the sorption of atrazine. Although the effect of organic C is removed as a factor contributing to variability, there are still clearly discernable areas within the field that have lower K_{oc} values relative to other areas. The largest of these areas has a K_{oc} of <125 and is located near the center of the field (X = 150, Y = 125). The K_{oc} values in this field are similar to other K_{oc} values reported for atrazine, which range from 90 to 300 in a wide range of soils (Montgomery, 1993), but are greater than the commonly used average K_{oc} of 100 reported in the SCS/ARS/CES database (Wauchope et al., 1992).

Landscape Position and Soils on Atrazine Sorption

Soil map units and the landscape position classes are shown in Fig. 1A and B. There is a close relationship between soil series and landscape position. Shoulder positions map almost exclusively as Clarion soils. The highest point in the field (X = 0, Y = 175) is in the shoulder area along the western edge of the site. Back slope positions are generally mapped as Clarion, Canisteo, and Harps soils. Soils in the closed depressions forming the large pothole (X = 175, Y = 175) and the small pothole in the southeast corner of the field are mapped mainly as Okoboji and Canisteo soils. The level upland area in the southwest quarter of the field is a swale area that lies between land draining into the large pothole and land draining to the southwest, and this area is mapped as Harps and Canisteo soils.

The soils in the upland shoulders sorbed the least atrazine of all landscape positions (Table 2). Mean K_d values of back slope positions were also smaller than the K_d of the depression soils. The K_d values measured in soils from the other landscape positions are intermediate between the shoulder and depression soils. The

Table 2. Atrazine sorpiton coefficients and soil properties expressed as pooled means of block-kriged values among landscape position classes within a 6.25-ha field.†

Landscape position	Atrazine K_d	Atrazine K_{oc}	Organic C	Clay content	pH
	L kg^{-1}	L kg C^{-1}	%	%	
Shoulder n = 15	3.2 ± 1.5	181 ± 48	1.7 ± 0.3	27.9 ± 0.7	5.9 ± 0.4
Back slope n = 60	4.6 ± 2.1	174 ± 40	2.7 ± 0.8	29.8 ± 1.9	6.3 ± 0.8
Foot slope n = 21	5.2 ± 2.2	181 ± 25	2.8 ± 0.5	31.9 ± 1.2	6.1 ± 0.6
Level upland n = 12	5.9 ± 1.2	193 ± 28	3.1 ± 0.4	42.4 ± 1.3	6.3 ± 0.6
Depression n = 13	8.4 ± 3.1	187 ± 41	4.3 ± 0.7	41.4 ± 2.4	6.8 ± 0.4
Whole field n = 121	5.1 ± 1.8	179 ± 38	2.8 ± 0.9	32 ± 9.1	6.3 ± 0.7

† Values are pooled means and standard deviations from block kriging over 25- by 25-m blocks.

Table 3. Atrazine sorption coefficients and soil properties among soil series within a 6.25-ha field.†

Soil series	Atrazine K_d	Atrazine K_{oc}	Organic C	Clay content	pH
	L kg^{-1}	L kg C^{-1}	%		
Clarion $n = 38$	3.8 ± 2.2	188 ± 43	2.0 ± 0.5	28 ± 1.2	5.8 ± 0.3
Nicollet $n = 2$	4.7 ± 3.1	223 ± 79	2.0 ± 0.03	27 ± 0.2	5.5 ± 0.4
Canisteo $n = 41$	6.0 ± 3.2	192 ± 28	3.0 ± 0.7	37 ± 1.7	6.3 ± 0.5
Harps $n = 14$	4.9 ± 1.4	152 ± 30	3.3 ± 0.4	35 ± 2.5	6.9 ± 0.6
Okoboji $n = 11$	7.0 ± 2.5	153 ± 18	4.5 ± 0.7	32 ± 2.9	7.3 ± 0.3
Whole field $n = 106$	5.1 ± 1.8	179 ± 38	2.8 ± 0.9	32 ± 9.1	6.3 ± 0.7

† Values are pooled means and standard deviations from block kriging over 25- by 25-m blocks. Only blocks containing at least 66% of a single soil type were used in the analyses; 15 blocks did not meet this criteria.

trend towards increasing organic C and clay contents in lower landscape positions (Table 2) appears to account for the increase in atrazine sorption by soils in these areas. Estimates of K_d are as variable within landscape position classes as within the entire field.

Areas with low atrazine sorption (Fig. 3) are consistently mapped as Clarion soils. The mean K_d and organic C content of the Clarion soil are generally lower than K_d values for all other soils, except Nicollet (Table 3). There was substantial variation in block K_d values within the soil series (compare Fig. 1B with Fig. 3), resulting in standard deviations for map units that are as large as that for the entire field (Table 3). This is likely due to unmapped inclusions within the map units, such as the effect of the small pothole in the southeast corner of the field. This area is mapped as a Canisteo soil, but has K_d values exceeding 8.0, which are greater than K_d of the other Canisteo soils in the field. There appears to be little difference between the Harps, Canisteo, and Nicollet soils in their sorption of atrazine, although this site included only a small sampling of Nicollet soils. Okoboji soils sorbed more atrazine than the other soil series, which is consistent with their greater organic C contents (Table 3).

Predicting Sorption at the Field Scale

Sorption of herbicides to soil is one of the more significant factors affecting pesticide leaching. Field-scale estimates of the potential leaching of herbicides require spatially distributed estimates of herbicide sorption. Intensive sampling coupled with geostatistical analysis can produce high resolution descriptions of the spatial distribution of sorption, but the resources necessary for high intensity sampling may limit the area that can be sampled. Maps of soil taxonomic units allow large heterogeneous areas to be subdivided into smaller, more uniform areas (strata), which can be sampled less intensively (Webster, 1985). Soil survey maps and associated chemical and physical data have been used to predict pesticide leaching at the field scale (Persicani, 1995) and in larger-scale assessments (Khan and Liang, 1989; Loague et al., 1990). In these studies, herbicide sorption within soil taxonomic units was estimated from soil survey information on organic C contents and published K_{oc} values. However, these large-scale studies do not assess whether local variations in soil or landscape features exist that result in greater potential leaching. Landscape position classes offer an alternative to soil map units for the basis of a sampling and/or estimation procedure. Thus, estimated or measured K_d values for the landscape positions or soil series can be used to produce spatial estimates of sorption over larger areas.

We used intensive sampling and geostatistical methods to produce two descriptions of atrazine sorption in surface soils at this site (Fig. 3 and 4). For comparison, maps of the site were produced by assigning the appropriate pooled mean for a landscape unit or soil series (Tables 1 and 2) to each of the corresponding 121 (25 by 25 m) blocks shown in Fig. 4. Assuming that the pooled means of atrazine K_d for soil series and landscape positions are similar to those that would have been obtained by a stratified random sampling of these units, then the maps of atrazine sorption based on the pooled means (Fig. 6 and 7) represent the spatial distribution of K_d that would be obtained using less intensive sampling methods. Comparison of the kriged map of atrazine sorption (Fig. 4) to maps based on the county soil survey (Fig. 7) or landscape position analysis (Fig. 6) suggests that landscape position was more accurate in representing the spatial variations within the field. We confirmed this by regression of the kriged K_d estimates for the 121 blocks against the corresponding estimates for these blocks predicted by soil series or landscape position. The resulting r^2 for the regressions was 0.53 and 0.72, respectively. Both of these methods appear to reduce the overall variability within the field, as would be expected from a means-based prediction scheme. Our re-

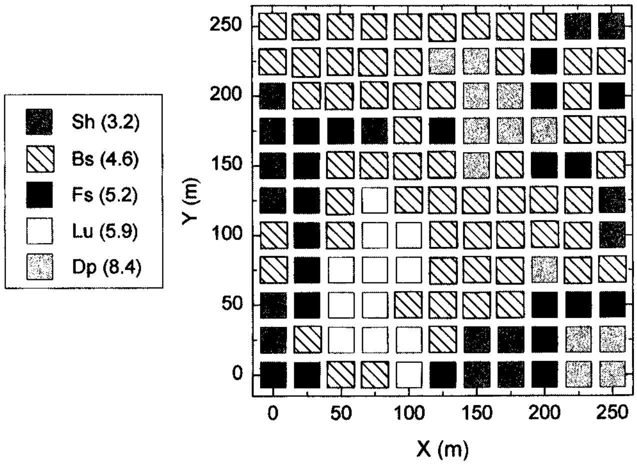

Fig. 6. Contour map based on mean atrazine K_d for landscape shoulder (Sh), back slope (Bs), foot slope (Fs), level upland (Lu), and depression (Dp) landscape positions. Values in parentheses are pooled means of block-kriged K_d estimates for all blocks within a landscape position. The appropriate pooled means were then reassigned to the 121 blocks to create the map.

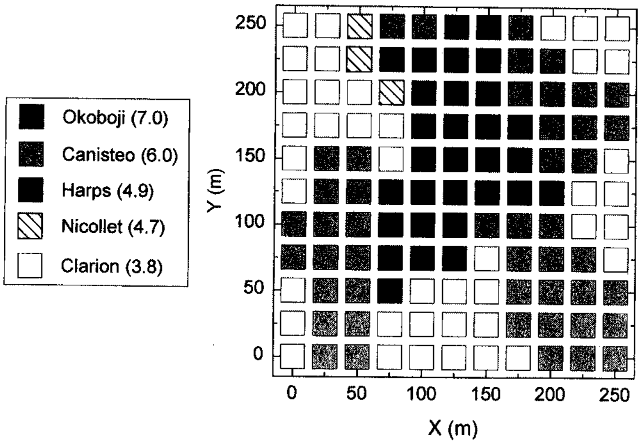

Fig. 7. Distribution of atrazine K_d based on the mapped units of five soil series (Clarion, Canisteo, Harps, Nicollet, and Okoboji). Pooled means of atrazine K_d values for each soil series were calculated from block-kriged estimates of K_d for all blocks mapped to a soil series. The appropriate pooled means (values in parentheses) were reassigned to the 121 blocks to create the map.

sults suggest that less intensive stratified sampling of soil series or landscape positions would produce reasonable representations of the variation in atrazine sorption at the field scale. Sampling of landscape position classes and soil series over a broader area will be required to fully evaluate the effectiveness of this approach at larger scales. In addition, the estimation of sorption is only one part of the process in estimating potential leaching.

Implications for Herbicide Transport

The larger K_d values in the depressional areas of this field may be an important factor affecting atrazine leaching and loss at this site. The depressional areas have increased water input relative to other areas in the field due to runoff from the upland areas. Like many of the depressional areas in this region, the large pothole in this field is tile drained. Atrazine concentrations in tile drainage water leaving this field in 1992 were generally below 2 $\mu g\ L^{-1}$ and accounted for approximately 0.045% of the applied herbicide (Jaynes et al., 1994). Other studies on a Nicollet soil at a nearby site showed similar results, although 40% of the tile-drained water samples had concentrations exceeding 3 $\mu g\ L^{-1}$ of atrazine (Jayachandran et al., 1994). As this water percolates through the profile to the tile drain, the larger sorptive capacity of the Harps and Okoboji soils tend to retard atrazine movement. Greater retention due to adsorption allows greater opportunity for atrazine biodegradation, which may lessen the total atrazine load in water leaving the field. This suggests that drainage tile depth and conservation of soil organic C are practices that would improve water quality relative to pesticides.

ACKNOWLEDGMENTS

We thank Elizabeth A. Douglass, Jody Ohmacht, Jennifer Moscinski, and Donna Schmitz for their technical advice and assistance.

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