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A simple index explains annual atrazine transport from surface runoff-prone watersheds in the north-central USA

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Abstract:

Year-to-year dynamics in weather affect both the timing of application and the potential hydrologic transport of pesticides. Further, the most commonly used pesticides dissipate in the environment during the growing season. Interactions among these factors – hydrology, timing of application and dissipation kinetics – hinder the detection of temporal trends in transport. It is increasingly important to be able to discern such trends, to judge effectiveness of management practices or to determine whether observed changes were caused by management or weather. In previous work, a cumulative vulnerability index was developed to account for these three factors. It explained 63% of annual variation in atrazine load in the Goodwater Creek Experimental Watershed (GCEW). The objectives of the current work were (i) to generalize the cumulative vulnerability index to explicitly account for variation in watershed size, area treated with atrazine and average application rate; (ii) to test the overall performance on watersheds showing such variation; and (3) to test whether the generalized index properly accounted for the additional input parameters. The generalized index was tested using data from GCEW (73.7 km^2) and seven additional watersheds in the northeast Missouri claypan region that varied in size from 212 to 1180 km^2 and from 4% to 23% of watershed area planted to corn or sorghum. Across 32 site-years, the generalized index explained 84% of variation in annual atrazine load. Further, tests of residuals showed no dependence on either watershed area or fraction of area planted to corn and sorghum, indicating that these parameters were properly integrated into the index. The performance of the index supports the conclusion that data obtained from GCEW is representative of the Mark Twain Lake Basin and likely the entire Central Claypan Major Land Resource Area. Published 2012. This article is a U.S. Government work and is in the public domain in the USA.

KEY WORDS atrazine; vulnerability index; watershed; claypan; CEAP; Goodwater Creek Experimental Watershed

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INTRODUCTION

Despite the obvious value to managers and policy makers that quantitative knowledge of contaminant transport would represent, few options exist to provide the information. Sampling and analysis costs are high, and it is not always apparent whether information obtained in one watershed is transferable to others with different land use, basin area or spatial variation in weather. Process-level models (e.g. the Soil Water Assessment Tool; Gassman *et al.*, 2007) have been built to account for the known mechanisms and processes, but mixed model performance results, high input requirements and high user training needs reduce the utility of the approach. Several attempts have been made to apply multiple linear regression models, but we know of few successes, primarily because of the difficulties listed in the following paragraphs.

Estimating contaminant transport in runoff in either forecast or hindcast (i.e. retrospective) mode is complicated by the dynamics of the driving forces, mitigating or exacerbating effects of antecedent and current conditions, and the transformations or dissipation kinetics of contaminants. The primary driver for runoff is precipitation, which is highly variable in space and time regarding frequency, intensity and event total. Further, antecedent rainfall, runoff, drainage and evapotranspiration fluxes integrate to the soil water balance, which controls the storage capacity of the soil. Yet another complication is the possibility that runoff could be initiated by either rain totals exceeding soil storage capacity or rain intensity exceeding infiltration rate. The result of these considerations is that runoff data are frequently more dynamic than the driving factor, rain, itself quite dynamic. These issues would complicate a calculation of a conservative tracer.

When the contaminant in question is a soil residual herbicide, two additional dynamics must be considered. Herbicide efficacy depends on the material being present in an active form long enough to suppress weeds, yet environmental and safety concerns require low to moderate persistence in soil. Most have dissipation kinetics that can be modelled as a declining balance of some kind, with the residual amount very small after some identifiable period. The second dynamic relates to when the material is applied to the field. For single fields, the timing of application is often known, and dissipation kinetics can be observed and modelled. For example, edge of plot and edge of field declines in atrazine concentrations were shown to follow first-order kinetics with nearly the same coefficients (Ghidey *et al.*, 2005, 2010). In larger watersheds that include many fields, herbicide application timing is affected by weather, soil moisture conditions and availability of equipment and labour to do the work. For many important soil residual

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herbicides, the timing is as near that of planting as possible (or coincident) to maximize weed control while the crop is short and not shading the weed seedlings. However, both herbicide application and planting require sufficiently dry soil to support the operation. Labour and equipment constraints may result in delayed or early herbicide application compared with the optimum time. As larger areas are considered, there may be considerable spread in time between the first and the last application of an herbicide during any given planting season, with precipitation and runoff often occurring after the first field is treated and before the last field is treated. Thus, interactions between the timing of application and the timing of runoff further increase the dynamic nature of contaminant transport.

Lerch *et al.* (2011a) provided data for atrazine transport from the Goodwater Creek Experimental Watershed (GCEW), in which a 15-year record of annual transport ranged from ~10 to ~280 kg year^{-1} (Figure 1). They found no independent variable that was correlated with annual transport. It closely matched the second-quarter runoff, but as runoff is used with concentration to calculate load, that correlation could not be used for explanatory purposes. Further, there were no trends discernable from the highly dynamic atrazine loads. To help explain the observed variation in load, they developed a cumulative vulnerability index (CVI) that explained 63% of annual variation in the load using a combination of three factors: timing of application, flow, and dissipation kinetics. As they were operating within a single watershed, the watershed area was not a factor, and the land use and the herbicide use in the watershed were fairly uniform during the period. Thus, the CVI as they developed it was functional in only that watershed. It remained to be seen if the CVI could be generalized to other watersheds of different areas and land use distributions.

A recent data set provided the opportunity to examine whether the original index could be generalized. In 2003, several US Department of Agriculture (USDA)–Agricultural Research Service (ARS) locations initiated a Watershed Assessment Study (WAS) within the Conservation Effects Assessment Project (CEAP), in partnership with USDA–Natural Resource Conservation Service (NRCS) and several other cooperating Federal and State agencies. The overall goal of CEAP was to determine the effectiveness of conservation practices at watershed scale to help inform the development of the next Farm Bill. Within the ARS CEAP-WAS in Missouri, the Cropping Systems and Water Quality Research Unit conducted a sampling campaign in several watersheds that drain into the Mark Twain Lake in northeast Missouri. The data produced in the Missouri ARS CEAP-WAS enabled this research.

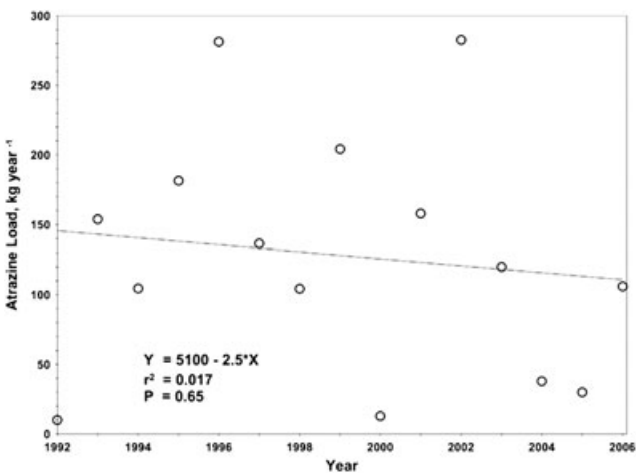

Figure 1. Atrazine load for 1992–2006 from the long-term ARS GCEW (Lerch *et al.*, 2011a)

The objectives of the current paper were (i) to generalize the CVI of Lerch *et al.* (2011a) to explicitly account for varying watershed area, varying area planted to corn and sorghum, and average application rate; (ii) to test the overall performance on watersheds showing such variation; and (iii) to test whether the generalized index properly accounted for the additional input parameters.

MATERIALS AND METHODS

Flow measurements

Year-round flow measurements were obtained from eight locations listed in Table I and shown in Figure 2. The USDA-ARS GCEW was instrumented for discharge and precipitation in 1971 (Sadler *et al.*, 2006). It has a 5:1 broad-crested V-notch weir for low-flow measurement and drains 73.7 km^2. The other seven sites are the US Geological Survey (USGS) flow stations (http://waterdata.usgs.gov/nwis/sw, access confirmed 15 August 2012) and drain areas ranging from 212 to 1180 km^2. Daily flow data for 2006–2009 were obtained for all sites.

Water sampling

An automated sampler (ISCO* 3230 and later 4230 Flowmeters; Teledyne Isco, Lincoln, NE, USA) was installed at the Goodwater Creek site in 1992 and programmed to take flow-proportional samples when the stage exceeded 0.15 m. These samples were supplemented during baseflow conditions by weekly grab samples (Lerch *et al.*, 2011a). In 2005, automated samplers (Sigma 900MAX; America Sigma, Inc., New York, NY) were installed at the USGS stations by USDA-ARS and operated nominally from April through November each year, with programming to take flow-proportional samples when stage rose approximately ≥ 0.15 m. These samplers were adjusted to follow the stage during baseflow conditions and thus sample small events on these larger rivers. Samples were composited into 10-l bottles. During the winter, grab samples were taken biweekly if not frozen, or if the water could be accessed through any ice that had formed. This report includes atrazine concentration and stream discharge data from the eight sites described previously for 2006 through 2009.

Chemical analysis and calculations

In the laboratory, samples were filtered through 0.45-μm nylon filters within 48 h of receipt, and herbicides were

Table I. Watershed area (km^2) and values for area (C + S area, km^2) and fraction (C + S fraction, dimensionless) of the watershed planted to corn and sorghum and annual atrazine loads (kg) for eight watersheds during the period 2006–2009

Site	USGS ID	Year→ Watershed area (km^2)	2006 C + S area (km^2)	2006 C + S fraction	2006 Load (kg)	2007 C + S area (km^2)	2007 C + S fraction	2007 Load (kg)	2008 C + S area (km^2)	2008 C + S fraction	2008 Load (kg)	2009 C + S area (km^2)	2009 C + S fraction	2009 Load (kg)
Goodwater Creek	NA	73.7*	14.5	0.20	83	15.2	0.21	60	8.6	0.12	190	16.0	0.22	180
Long Branch (lower)	5506100	480	95	0.20	390	106	0.22	390	74	0.15	1100	110	0.23	710
South Fork Salt River	5504800	589	77	0.13	200	83	0.14	220	68	0.11	690	87	0.15	930
Lick Creek	5507600	260	49	0.19	36	60	0.23	53	43	0.17	570	56	0.22	750
Elk Fork	5506800	517	42	0.08	240	45	0.09	260	32	0.06	520	56	0.11	400
Middle Fork	5506350	865	44	0.05	250	42	0.05	160	34	0.04	560	58	0.07	630
Crooked Creek	5503800	212	29	0.14	84	26	0.12	69	25	0.12	210	30	0.14	290
North Fork	5502500	1180	68	0.06	235	73	0.06	380	49	0.04	1200	71	0.06	1400
Totals, excluding Goodwater Creek**		4100	403		1440	436		1530	325		4850	468		5110

*Lerch *et al.* (2011a) reported the historical area of 72.5 km^2; values in this table were derived using a Digital Elevation Model based algorithm, producing a difference of 1.6%.
**Goodwater Creek is nested within the Long Branch watershed; thus, area and load are both included in the Long Branch values. Sums are rounded.

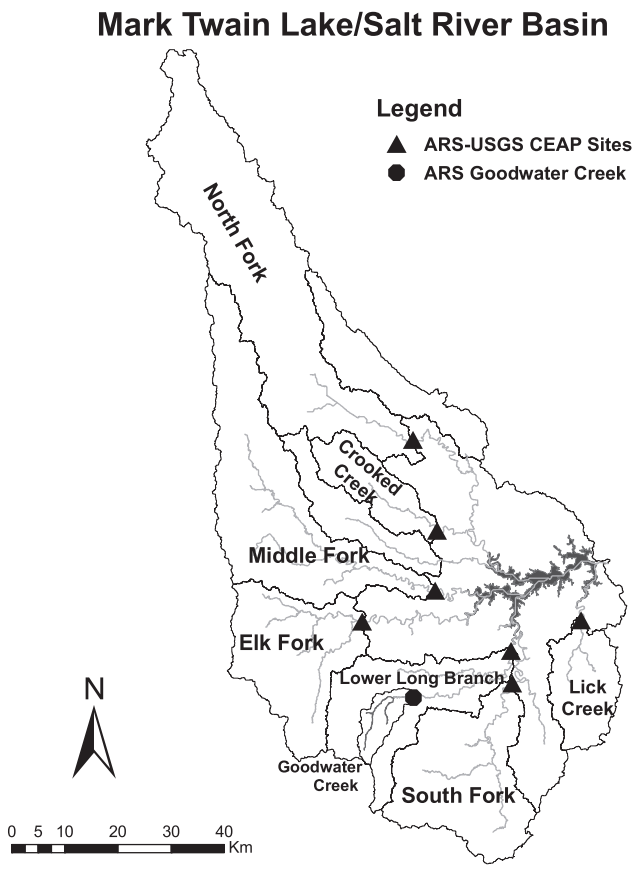

Figure 2. Sampling sites in the Mark Twain Lake/Salt River Basin, consisting of seven USGS flow stations augmented with samplers and the long-term ARS GCEW

extracted with C_{18} solid-phase extraction cartridges using 200 ml samples. The solid-phase extraction cartridges were eluted with ethyl acetate and concentrated ~600-fold under a stream of ultra pure N_2. Atrazine concentrations were determined by gas chromatography–mass spectrometry using a Saturn 2000 ion-trap mass spectrometry detector (Varian Inc., Harbor City, CA). The limit of detection was 0.003 $\mu g\ l^{-1}$. Details of the method were reported by Lerch and Blanchard (2003).

Sample numbers varied annually and across sites as a result of annual and spatial variation in rainfall that caused runoff events. At the USGS sites, the number averaged from 32 to 42 samples per year, with an absolute minimum of 20 at Lick Creek during 2007 and a maximum of 52 at Lower Long Branch during 2008. Concentrations were interpolated to daily values, which were multiplied by daily flow volumes to obtain daily load.

Cropland area

Annual areas planted to corn and grain sorghum were obtained from the National Agricultural Statistics Survey (NASS) Cropland Data Layer (CDL; USDA-NASS, 2010). The 56-m resolution CDL is created annually by NASS using multispectral imagery from Resourcesat-1 AWiFS, ground truth training data from the USDA Farm Services Agency and various ancillary data sets (Mueller *et al.*, 2009). Using ArcGIS® (ESRI, Redlands, CA), CDL data

sets for each year were overlain with watershed boundaries, and the area within each watershed in corn, grain sorghum and other land uses was determined. An example for GCEW in 2009 is given in Figure 3.

Timing of application

Daily planting progress was used as a surrogate for herbicide application timing, and these data were obtained from weekly planting progress data for the northeastern Missouri crop reporting district (USDA-NASS, 1992-2009). They are shown in Figure 4 with the 1992–2005 data included to demonstrate the interannual variability in the field operations. In these data, the first nonzero value observed was the first value reported, and the last value reported was often not 100%, as the planted acreage was expressed as a fraction of the expected planted area. Thus, a zero was inserted one week ahead of the first nonzero value, and all values were scaled to the maximum planting progress, making a range of weekly values from 0 to 1. This series of weekly values was then expanded to daily values by linear interpolation, and the daily planting fraction for each day, j (DP_j), was found by difference from the previous day. The first day, last day and length of the planting season were also obtained from that sequence.

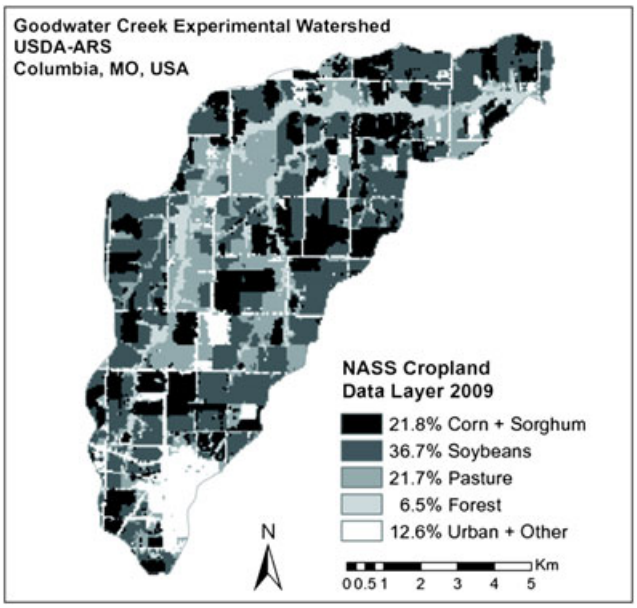

Figure 3. Example of NASS Cropland Data Layer used to determine intensity of cropping. (Data obtained from NASS, http://www.nass.usda.gov/research/Cropland/SARS1a.htm)

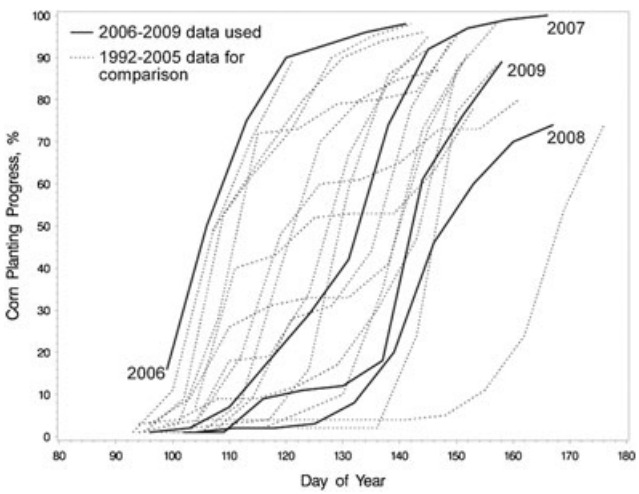

Figure 4. Northeast Missouri district-level corn planting progress for 1992–2009. The planting data in the current work are shown using solid lines; the broken lines illustrate the timing relative to the prior years. (Data obtained from NASS, http://www.nass.usda.gov/Statistics_by_State/Missouri/Publications/Crop_Progress_and_Condition/)

Atrazine dissipation kinetics

The original CVI (CVI_o) had been developed using the edge-of-plot results of Ghidey *et al.* (2005) in which dissipation kinetics for concentration from mulch-till corn plots were best described as a first-order kinetics process $e^{(-kt)}$ using $k = 0.0625$ day^{-1}. Ghidey *et al.* (2005) also showed the same decline applied for load. Since then, Ghidey *et al.* (2010) have demonstrated the equation successfully scales up 100 times from the 0.34-ha plots reported in 2005 to a 35-ha field reported in the later paper. The best fit for the field data produced $k = 0.0612$ day^{-1} (Figure 5). As the current work is attempting to integrate multiple fields to watersheds, the field-edge value was used to develop the generalized index.

Original CVI

The CVI developed by Lerch *et al.* (2011a) was based on two equations, one for computing daily weights (Equation 1) and the other for computing the CVI for a given year (Equation 2). Equation 1 accounted for timing after application without involving quantitative discharge data by reducing the hydrograph to a series of binary event indicators,

$$DW_i = \sum_{i=1}^{LA} Ev_i \times e^{(-kt)} \quad (1)$$

where DW_i was the daily weight; Ev_i was the daily value of the event indicators, set equal to 0 if the daily discharge was <10 mm day^{-1} and equal to 1 if daily discharge

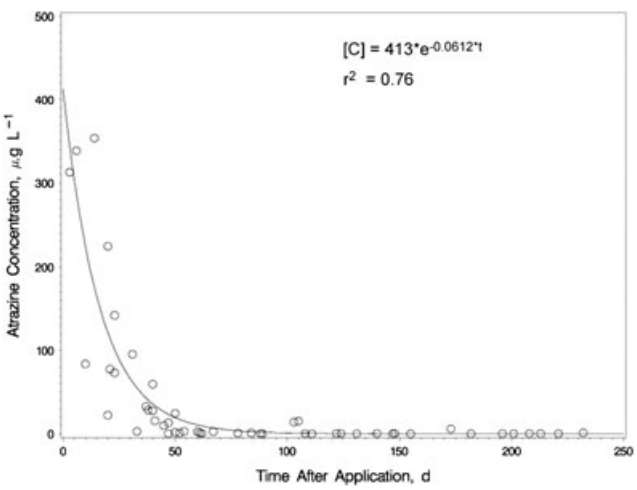

Figure 5. Edge-of-field (35 ha) atrazine concentrations were demonstrated to dissipate exponentially with $k = 0.0612$/day by Ghidey *et al.* (2010)

was >10 mm day^{-1}; k was the first-order rate constant for atrazine dissipation kinetics, set equal to 0.0625 day^{-1} based on edge-of-plot data (0.34-ha plots) from Ghidey *et al.* (2005); t was time since application, in days; and LA was the length of time over which the daily weights were computed after each daily application, chosen to be 100 days and with the assumption that planting time is a surrogate for application time. This corresponded to a minimum weight of 0.000335 using $k = 0.0625$ day^{-1}. Equation 2 computed the annual index weight for any particular year as given by,

$$\mathrm{CVI_o} = \sum_{j=1}^{\mathrm{LS}} \mathrm{DW}_j \times \mathrm{DP}_j \tag{2}$$

where $\mathrm{CVI_o}$ was the original cumulative vulnerability index, DW_j was the daily weight computed from Equation 1, DP_j was the daily planting progress fraction and LS was the length of the planting season for a given year. The time series of planting progress and runoff events started the first day of planting and was extended 100 days beyond the last day of planting. The annual $\mathrm{CVI_o}$ for each of the 15 years of the study was correlated to atrazine load and shown to explain 63% of the variation in annual atrazine load. Combining the previous equations gives Equation 3:

$$\mathrm{CVI_o} = \sum_{j=1}^{\mathrm{LS}} DP_j \times \sum_{i=1}^{\mathrm{LA}} Ev_i \times e^{(-\mathrm{kt})} \tag{3}$$

Because $\mathrm{CVI_o}$ was developed specifically for a single watershed (GCEW) with constant area and somewhat consistent area treated with atrazine, the simple regression above worked fairly well for both the 1992–2006 development and the 2007–2008 validation data.

Generalized CVI

However, to be applicable across watersheds of varying area and pesticide use intensity, we expected that these parameters would be explicitly required. This was confirmed (data not shown) when the regression explained less than half as much variation in the 2006–2009 data set from all eight watersheds as in the original Lerch *et al.* (2011a) data set from Goodwater Creek.

In the generalized CVI ($\mathrm{CVI_g}$) Equation 4, the area planted to corn and sorghum (A_{cs}), the area of the watershed (A_{ws}) and the atrazine application rate (R, assumed constant at 1.63 kg ha^{-1} for this 4-year period per Murphy *et al.*, 2008) are explicitly included. Further, the watershed area was considered by testing the $\mathrm{CVI_g}$ against annual unit area load instead of annual mass transported. Recall that the edge-of-field decay constant of $k = 0.0612$ (Ghidey *et al.*, 2010) was used instead of the original edge-of-plot value of $k = 0.0625$ (Ghidey *et al.* (2005).

$$\mathrm{CVI_g} = \frac{A_{\mathrm{CS}} \times R}{A_{\mathrm{WS}}} \sum_{j=1}^{\mathrm{LS}} \mathrm{DP}_j \times \sum_{i=1}^{\mathrm{LA}} \mathrm{Ev}_i \times e^{(-\mathrm{kt})} \tag{4}$$

The overall fit of the generalized index was tested by regression of annual unit area load, most easily expressed in grams per hectare, against the $\mathrm{CVI_g}$. Acceptable performance could be concluded if the regression explained a comparable amount of the temporal and spatial variation as the $\mathrm{CVI_o}$ with the GCEW data set. A second test was to examine robustness of the residual data set against dependence on the watershed area and fraction of the watershed in corn and sorghum. This test was expected to be acceptable if the slopes of the regressions were not significant. If the index could be shown to be independent of the two parameters, it would be considered support for the index properly accounting for the two parameters.

RESULTS AND DISCUSSION

The primary test of whether the $\mathrm{CVI_g}$ performed well was whether annual variation in unit area load, in grams per hectare, could be explained. The goodness of fit demonstrated across the eight watersheds was unexpectedly good. It was superior to that of the $\mathrm{CVI_o}$ development data set, explaining 84% of the variation in unit area load for the eight watersheds during 2006–2009 (Figure 6). Compared with long-term average precipitation and runoff, this period included two fairly dry and two quite wet years, with expected spatial variation of rainfall within years, and is therefore considered representative of a wide range of weather conditions. A shown in Table I, the watershed areas included ranged from GCEW at 73.7 km^2 to the North Fork of the Salt River at 1180 km^2. Given that the development of the index was done at the smallest of these scales (73.7 km^2), the index is considered scalable to at least 16 times (1180 km^2) that of development. It is also worth noting that the dissipation kinetics model used appears scalable from 0.34 to 120,000 ha, or more than five orders of magnitude.

The eight watersheds included from 4% to 23% of the area planted to corn and sorghum, with three generally less than 10%, two between 10% and 15% and three higher than 15%. Fitting a single, simple index across such a range in cropping intensity and obtaining performance as good as shown in Figure 6 suggests that the index reasonably accounts for the presumed pesticide use intensity during this

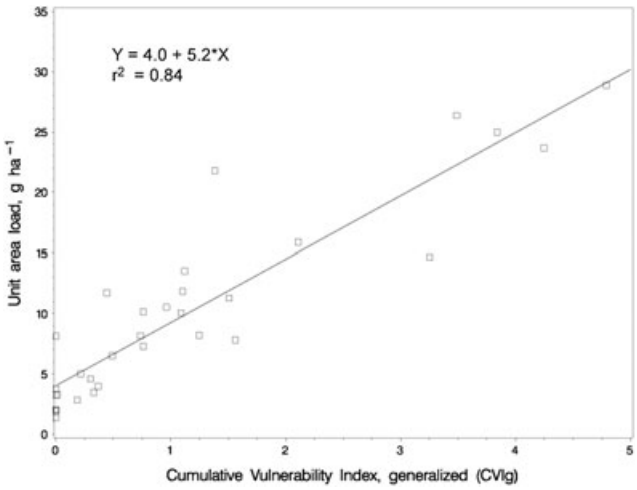

Figure 6. Plot of unit area atrazine load in grams per hectare against $\mathrm{CVI_g}$ for eight sites in the Mark Twain Lake basin, during 2006-2009

short period. A single average application rate was assumed for the 4-year test period, as no general trends were reported from observers of farm operations in the area. Thus, a rigorous test of the temporal or spatial variation in average application rate is not possible with these data. However, as the numerator of the index includes the product of area applied and the average application rate in that area, it would be expected that the index would properly account for that variable as well. From a practical standpoint, however, obtaining values for *R* may be so problematic at watershed scale to make average values, such as those obtained in farmer surveys (Murphy *et al*., 2008), more useful.

In a general sense, the CVI_g is consistent with other results, such as the Watershed Regressions for Pesticides model findings that watershed pesticide use intensity is a primary determinant of pesticide concentrations (Larson *et al*., 2004; Stone *et al*., 2008). However, where their index attempted to predict the frequency distribution of atrazine concentrations, averaged across years, this index explicitly accounts for temporal dynamics in timing of operations, hydrology and dissipation kinetics by assessing annual loads. Thus, it can be used as a retrospective tool to help interpret multiple-year data obtained in one or several watersheds.

A second test of model performance was whether or not the residuals from the regression showed any dependence on the input parameters, with the interpretation being that absence of dependence would support a conclusion that the index properly accounted for the parameters. The plot of residuals from regression line against watershed size is shown in Figure 7. The low slope and the low probability that the slope was significantly different from zero support the conclusion. The plot of residuals from the regression line against the watershed fractions in corn and sorghum is shown in Figure 8. Here, the slope is even less and the probability is even more likely that the slope is not different from zero. Thus, both tests support the conclusion that these two parameters are adequately accounted for.

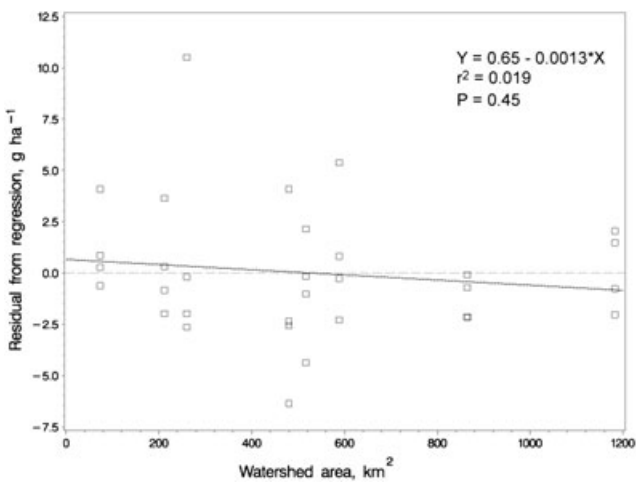

Figure 7. Plot of the residual from the regression in Figure 6 against watershed area. Probability stated is of the null hypothesis that the slope is zero and is clearly nonsignificant

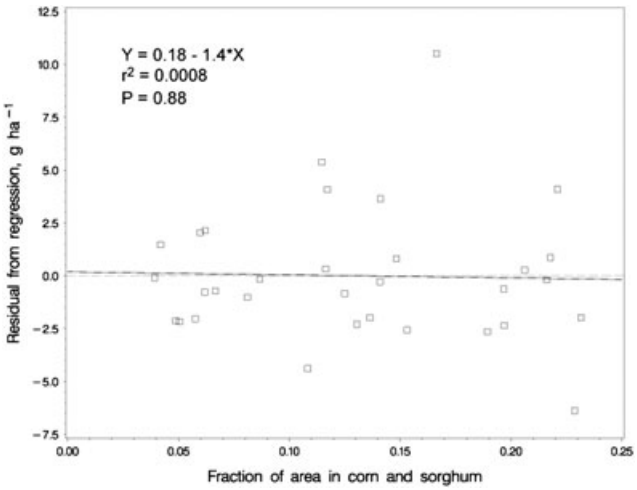

Figure 8. Plot of the residual from the regression in Figure 6 against fraction of the watershed in corn and sorghum. Probability stated is of the null hypothesis that the slope is zero and is clearly nonsignificant

Potential applications of CVI_g

One possible use of CVI_g is as a covariate in time series analyses to improve ability to detect trends in atrazine load. Preliminary analyses with the CVI_o are promising, and this will be examined rigorously in future research. Another possible use is to estimate the likely trends in atrazine load that would have been expected during years where no data were collected but that were important for regulatory concerns. One example could be the Mark Twain Lake, in Missouri, USA, which was on the original list of impaired waters for atrazine and removed in 2003 (USEPA, 2007a). At the time of the delisting, research conducted within the GCEW watershed raised the possibility the observed reductions in lake atrazine concentrations might have been caused by dry weather. This concern could be evaluated using the CVI_g if watershed-level cropping data could be developed. Developing those data would not be as simple as for the current work, as the NASS CDL was not available for that part of Missouri until 2006. However, with county-level data and correlations between it and the NASS CDL in these later years, this could be done. In addition, if the recent wetter weather raises concerns about atrazine impairment, this index could help both regulatory agencies and production interests interpret the data.

Knowing whether data obtained in one watershed can be transferred to another would be quite valuable to all parties affected by implications of the data. In fact, knowing if those data are representative, and over what geographic area they are, may be one of the most important functions of models and other technology transfer tools, including the CVI_g index. The goodness of fit obtained with CVI_g strongly supports that it is applicable to the entire Mark Twain Lake basin, which is some 6600 km^2. Similarities between the soils in the basin and the surrounding area suggest that it is also applicable to the entire Central Claypan Major Land Resource Area, MLRA 113 (USDA-NRCS, 2006). Certainly, there is little scientific basis for claiming differences in pesticide transport dynamics within MLRA 113. There could be arguments made that it is applicable to

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other surface runoff-prone watersheds outside the MLRA, but that interpretation is certainly not conclusive.

Another advantage of the approach used in developing the CVI_g is its applicability to other pesticides that are vulnerable to transport in surface runoff. For example, Ghidey *et al.* (2005) documented dissipation for metolachlor at edge of the same plots as for atrazine, and Ghidey *et al.* (2010) showed that, with an accounting for thermal effects, dissipation of metolachlor at the edge of the field could also be described. If the edge of plot or edge of field dissipation kinetics for transport in surface runoff are known, then the CVI_g should be applicable to any chosen pesticide compound, regardless of the kinetics (i.e. zero, first or second order), as long as the mechanism for transport is primarily surface runoff.

The key hydrologic feature of GCEW and watersheds throughout the MLRA 113 is the presence of a subsurface claypan with smectitic mineralogy. The claypan promotes surface runoff when it is at or near saturation by limiting the available water holding capacity to the thickness of the soil above this restrictive layer. Therefore, other soils with smectitic mineralogy and the presence of argillic horizons or fragipans that also serve as restrictive layers will have similar hydrology to that of claypan soils. This potentially extends the applicability of the CVI_g to other regions of the Midwest, including the following MLRAs: 106 (Nebraska and Kansas Loess-Drift Hills), 109 (Iowa and Missouri Heavy Till Plain), 112 (Cherokee Prairies) and 114 (Southern Illinois and Indiana Thin Loess and Till Plain, Western Part). Impaired water designations or other documentation of high atrazine concentrations have been identified for some watersheds within all of these areas (USEPA, 2007b and USEPA, 2010). This would extend the applicability of the CVI_g to more than 18 million ha.

However applicable it might be outside MLRA 113, the performance of the CVI_g in the watersheds studied is a persuasive argument that the atrazine transport data obtained in GCEW is generally applicable in the central claypan region. This conclusion is fully consistent with other studies conducted at much reduced data frequency within the northern Missouri/southern Iowa region in which Goodwater Creek or its larger basin, Long Branch Creek, were shown to be representative of the Claypan region and/or Salt River basin (Blanchard and Lerch, 2000; Lerch and Blanchard, 2003; Lerch *et al.*, 2011b). To our knowledge, the current research results are the strongest scientific basis for generalization within these regions.

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DISCLAIMER

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