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IRRIGATION METHODS AND TRANSPORT OF IMAZAMETHABENZ-METHYL TO GROUND WATER: GREENFIELDS BENCH, MONTANA[1]

Kathleen J. Miller[2]

ABSTRACT: A series of gravel terraces support a shallow aquifer that is the sole source of drinking water for three public water supplies and more than 400 private wells on the Greenfields Bench in west-central Montana. Farming practices on the Greenfields Bench include irrigation of malting barley and the yearly application of herbicides for the control of weeds. The most commonly used herbicide (imazamethabenz-methyl, U.S. trade name Assert®) has been found in the ground water on the Greenfields Bench. An experiment was conducted in 2000 and 2001 to characterize the transport of Assert and its acid metabolite to ground water under three irrigation methods: flood, wheel line sprinkler, and center pivot sprinkler. Results show that Assert concentrations in ground water are controlled by hydraulic loading rates of each irrigation method, Assert persistence in soil, hydraulic characteristics of the aquifer, and adsorption/desorption of Assert onto clay particles and organic matter.

(KEY TERMS: imazamethabenz-methyl; irrigation; ground water contamination; ground water management; ground water flow.)

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INTRODUCTION

Three public water supplies and more than 400 private residences obtain their drinking water from the Greenfields Bench aquifer, a topographically isolated bench of Cretaceous age overlain by a veneer (1 to 12 m thick) of gravel deposits. The aquifer receives about 70 percent of its recharge from on-farm irrigation, leaking irrigation canals, and ponded tailwater (Osborne *et al.*, 1983). Water levels undergo dramatic seasonal fluctuations that correspond to the onset of irrigation, demonstrating the direct hydraulic connection between surface water and ground water.

The herbicide Assert was first detected in ground water on the eastern edge of the Greenfields Bench (Figure 1) during routine monitoring conducted by the Montana Department of Agriculture (MDA) in 1996. Sampling efforts were expanded to other areas on the Bench to provide an initial assessment of Assert occurrence in ground water. At the same time, the Montana Bureau of Mines and Geology assisted the Town of Fairfield with source water protection boundary delineation as part of a demonstration project (Miller *et al.,* 1998). Assert, Assert metabolite, picloram, prometon, and clopyralid were detected in four of the town's wells and in numerous private wells in the surrounding area.

The Weed Science Society of America (WSSA, 1994) describes Assert as an imidazolinone compound that contains the active ingredient imazamethabenz-methyl. It is a post-emergent herbicide used for control of wild oats, mustards, and buckwheat in barley and wheat. After rapid absorption through foliage and roots, Assert is translocated through the plant, where inhibition of amino acid synthesis and plant cell division of growing roots and shoots occurs. The recommended application rate is .057 to 0.71 liters per hectare (L/ha) (WSSA, 1994). The application rate within this range is largely determined by the density of weed infestation. Application rates need to be high enough to provide good weed control and minimize weed resistance yet low enough to minimize crop production costs. Herbicide efficacy is tied to application at the correct weed growth stage. This optimal time of

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[2]Senior Research Professor, Montana Bureau of Mines and Geology (MBMG), Montana Tech of the University of Montana, 1300 West park Street, Butte, Montana 59701 (E-Mail: kmiller@mtech.edu).

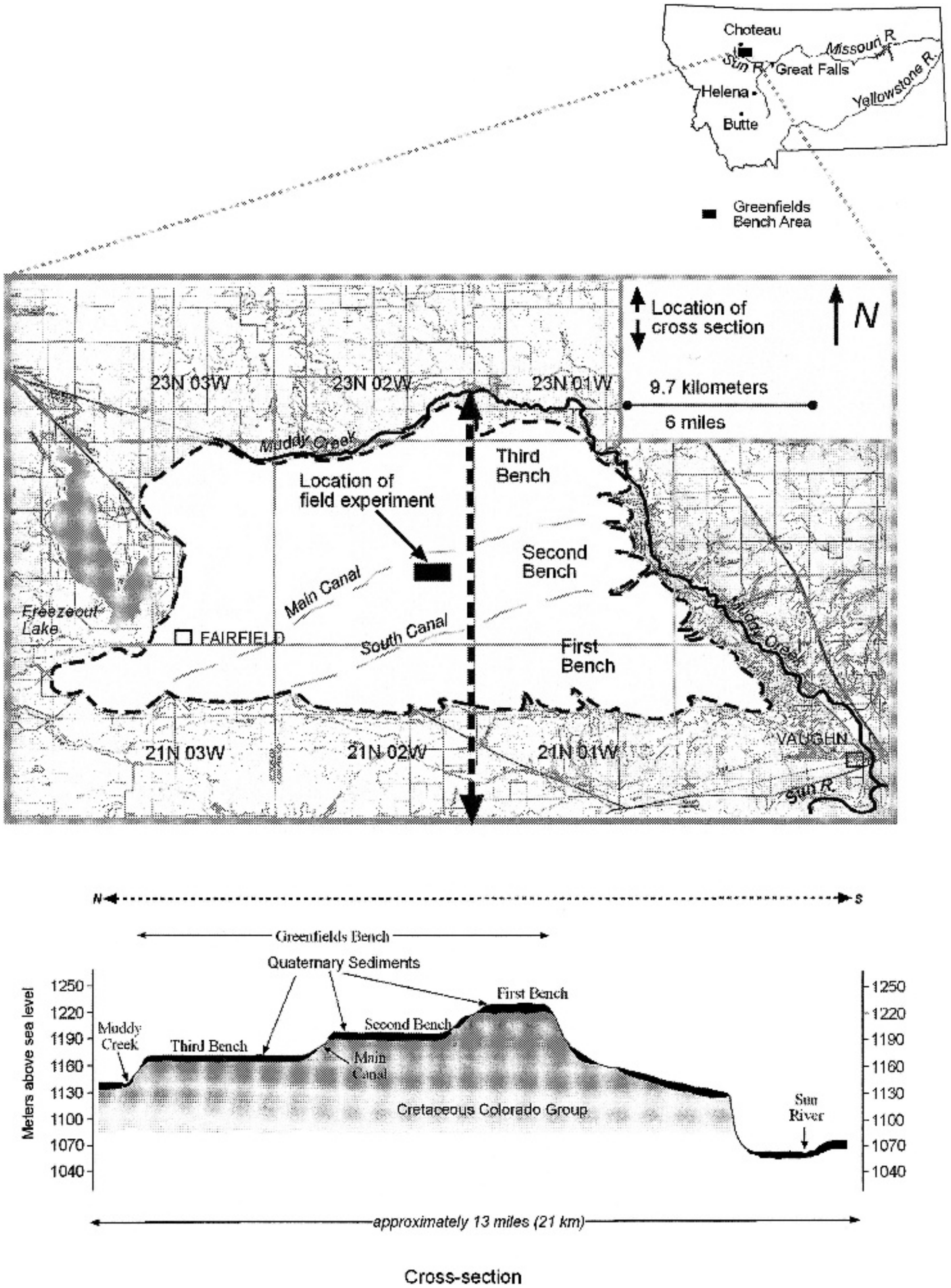


Figure 1. Project Location and Generalized Cross Section.

application often coincides with crop moisture needs and frequently results in crop irrigation shortly after Assert application. Assert is a 3:2 mixture of *para*-methyl and *meta*-methyl isomers with solubilities of 857 mg/L for the *para*-isomer and 1,370 mg/L for the *meta*-isomer. When applied, it undergoes hydrolysis. It hydrolyzes slowly at pH ranges from 5 to 7, but rapidly in alkaline conditions, resulting in the de-esterified imazamethabenz acid. It is the acid that provides the weed control (WSSA, 1994).

The U.S. Environmental Protection Agency (USEPA) Interim Health Advisory Level for Assert in drinking water is 400 micrograms per liter (µg/L). Although current data on Assert do not suggest any major toxicological problems, the chemical is extremely caustic and corrosive (USEPA, 1988), causing skin and eye irritation. The acceptable daily intake of Assert, based on a one-year dog feeding study and using a hundredfold uncertainty factor, is calculated to be 0.0625 mg/kg/day (USEPA, 1988).

In one of the few previously published investigations that address Assert concentrations in ground water, D'Ascenzo *et al.* (1998) reported that imazamethabenz-methyl was detected in seven samples collected near Celano, Italy, between June and September 1997 at concentrations ranging from 0.024 to 0.068 µg/L. Most previous investigators have described its persistence and mobility in soil. Wauchope *et al.* (1992) estimated the half-life of Assert in soil at 45 days. Pool (1993) noted that microbial decomposition and adsorption are important factors that affect Assert degradation in soil. He showed that its persistence is related to the content of clay and organic matter of soils. However, Assert is anionic, and therefore clay and organic matter account for minimal anionic adsorption. Pool observed that on its own, pH does not play a significant role in determining Assert activity, but noted that lowering the pH enhances sorption in soils. Rouchard and Gustin (1994) reported that at nonirrigated sites in Belgium, organic fertilizer treatments retarded the decomposition of Assert and its metabolites. Cartón *et al.* (1997), in a study of adsorption/desorption equilibria at different temperatures, noted fast initial adsorption for the first few hours followed by a phase of slow adsorption. Cartón *et al.* (1997) showed an important hysteresis phenomenon (nonreversible adsorption) that may affect imazamethabenz availability in soil. Photodecomposition (photolysis) of imadazolinone herbicides could be an important dissipation mechanism in the field. In Curran *et al.* (1992) ultraviolet light caused 87 percent degradation (photolysis) of imazamethabenz in aqueous solution after 48 hours.

In general, the processes that affect the transport of pesticides (inclusive of herbicides, fungicides, etc.) through the soil into ground water depend on the pesticide properties and environmental conditions. Pesticides that are not metabolized by the pest (weeds in this case) remain in the environment until they are degraded. Pesticides that are not rapidly degraded by microbial, photolytic, hydrolytic, or chemical processes and that have moderate to high solubility, poor sorption, and moderate to lengthy persistence will have the potential to leach to ground water.

The purpose of this study was to evaluate whether altering irrigation practices could reduce the occurrence of Assert in ground water. This report compares three commonly used irrigation methods and Assert residuals in soil and ground water to develop an understanding of the variables that affect aquifer recharge and mass flux. Assert is the target analyte in the experiment because of its extensive use on crops on the Greenfields Bench and because of its Bench wide presence in ground water. An observation well network and an intensive herbicide sampling strategy were used in a field experiment on a malting barley field to evaluate Assert concentrations in soil and ground water under irrigation by flooding, wheel line sprinkling, and center pivot sprinkling.

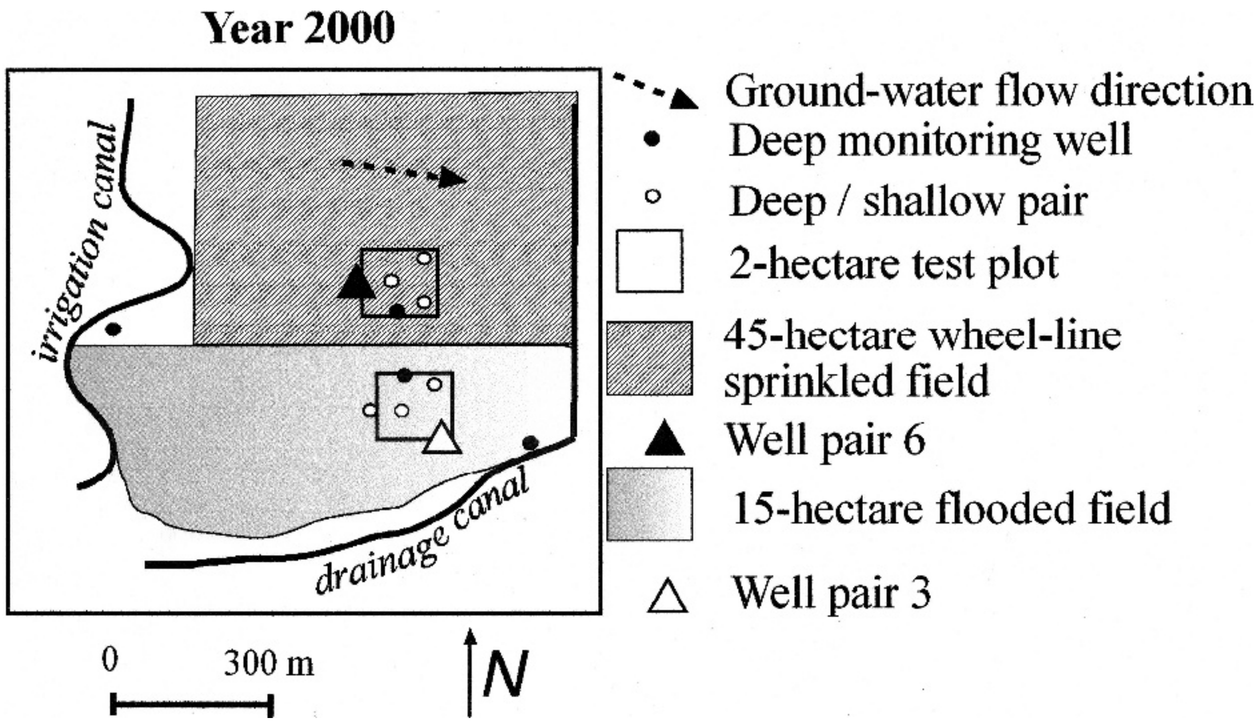
HYDROGEOLOGIC SETTING

The gravels of the Greenfields Bench are underlain by the Marias River Formation of the Cretaceous Colorado Group. The Marias River Formation constitutes approximately the upper 215 m of the 500 m thick Colorado Group and consists primarily of dark gray marine shale with some interbedded siltstone, sandstone, and bentonite (Maughan, 1961).

The top of the Marias River Formation is an erosional surface formed prior to the deposition of Tertiary/Quaternary age gravels, leaving a gently eastward sloping plain. Three terraces, the First, Second, and Third benches (Figure 1), are known collectively as the Greenfields Bench and were formed by the Sun River in pre-Wisconsin and early Wisconsin time during periods of downcutting and terrace gravel deposition. The present channels of Muddy Creek and the Sun River were formed later, probably during later Pleistocene time (Maughan, 1961). Across the Greenfields Bench, the gravel thickness ranges from 1 to 12 m and the soils are about 2 cm thick.

The gravel aquifer is the only source of drinking water for the residents on the Greenfields Bench and receives about 70 percent of its recharge from a combination of irrigation canal/ditch loss and on-field irrigation (Osborne *et al.,* 1983). Water level fluctuations of as much as 5.5 m in response to the onset of irrigation demonstrate the hydraulic connection between irrigation water and ground water. Ground water quality is characterized as a magnesium/bicarbonate type, with ground water pH ranging from 7.3 to 8.2. Average ground water alkalinity and total dissolved solids are 253 mg/L and 320 mg/L, respectively (Miller *et al.,* 2002). Ground water concentrations of Assert (or its acid metabolite) range from below the 0.2 µg/L limit of quantitation (LOQ) to 10.0 µg/L.

The study site is on the Second Bench (Figure 1). The saturated thickness of the aquifer at the experimental site is about 5.5 m during irrigation. Depth to ground water ranges from ground surface to 3 m below ground surface, depending on whether or not there is irrigation. The upper 1.8 m of the aquifer consists of poorly sorted gravel and sand embedded in a clay matrix, with the lower 3.6 m generally composed of well sorted sand and gravel. Soils on the study site are characterized as clay loam with pH 7.4 and 2.5 percent total organic carbon. Ground water flows in



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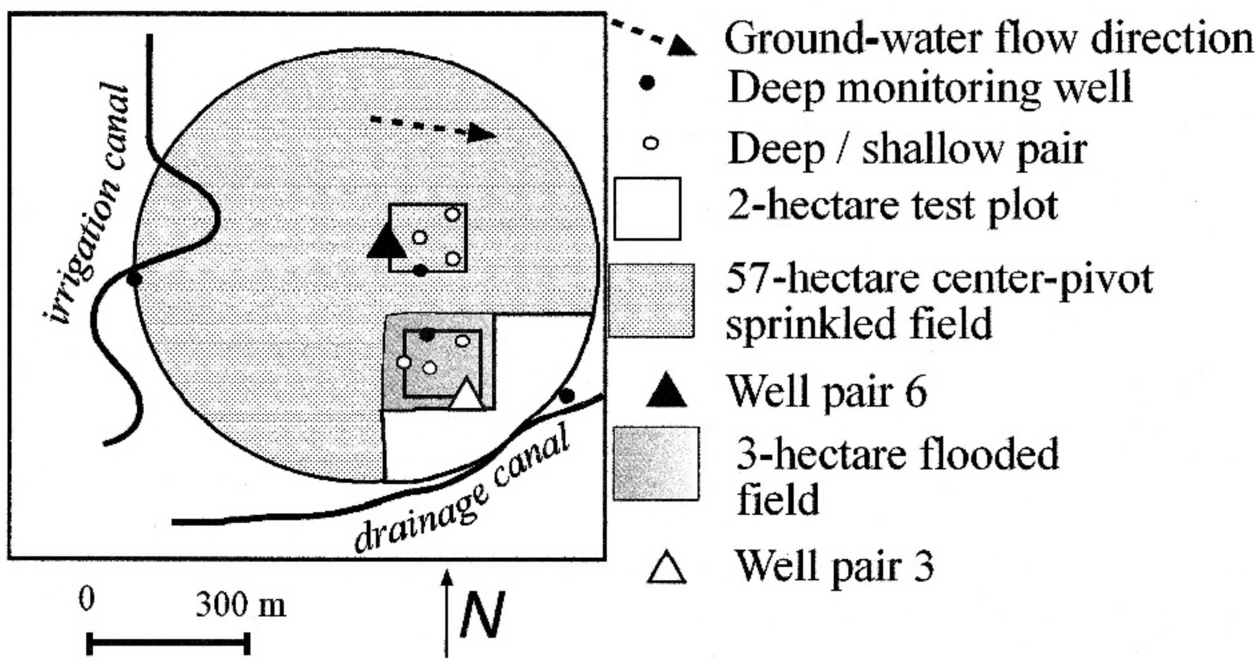


(b)

Figure 2. Schematic Map of Experimental Plots in (a) 2000 and (b) 2001.

an easterly direction on the study site (Figures 2a and 2b). Hydraulic conductivity is estimated at 180 m/day using results of short term aquifer tests performed on 13 wells across the Greenfields Bench. Hydraulic gradients range from 0.004 prior to irrigation to 0.008 across the flood irrigated plot (Miller *et al.,* 2002).

METHODS

The total field size, including both flood irrigated and sprinkle irrigated areas (Figures 2a and 2b), is 60 ha. One 2 ha test plot was located in the sprinkle irrigated field and another 2 ha test plot was located in the flood irrigated field. In 2000, the northern 45 ha (inclusive of one 2 ha sprinkler test plot) were irrigated with a wheel roll sprinkler and 15 ha (inclusive of the 2 ha flood test plot) were flood irrigated (Figure 2a). In the fall of 2001, the landowner replaced the wheel line system with a center pivot that is capable of irrigating the entire field. During the irrigation season of 2001, the landowner was able to interrupt the pivot cycle to avoid sprinkling the flood plot (Figure 2b). Only 3 ha (inclusive of the 2 ha flood test plot) were flood-irrigated in 2001, and 57 ha inclusive of the 2 ha sprinkler test plot were irrigated using a center pivot sprinkler system.

Twenty monitoring wells (Figures 2a and 2b), 16 of which were deep/shallow nested pairs, were installed using flush threaded PVC casing and screens with size 0.020 slots. Shallow wells were 3 m deep with screened intervals from 1.2 m to 3 m. Data collected from monitoring wells installed upgradient and downgradient of the test plots were used to characterize background water levels and Assert concentrations. Deep wells were 6 m in depth with screened intervals from 3 to 6 m. Shallow well data were compared with data from deep wells to evaluate the vertical distribution of Assert in the saturated zone. Unpreserved ground water and soil samples were analyzed by the Montana Department of Agriculture using MDA (2002) and American Cyanamid Co. (1991) methods. Random composite samples of the top 1.8 cm of soil were collected from each of the 2 ha flood and sprinkler plots and were composed of 15 random subsamples. Soil samples were collected prior to Assert application in 2000 and 2001 to determine textural characteristics and background Assert levels. Additional soil samples were collected at monthly intervals during the growing seasons. Ground water samples were collected prior to Assert application each year, then at weekly intervals for two months after Assert application. Assert was applied on June 10, 2000, and on May 31, 2001, at the rate of 1.8 L/ha.

Mass flux estimates, expressed in grams/day (g/day), were derived by multiplying Assert metabolite concentration values (µg/L) for shallow Wells 3 and 6 (Figures 3a and 3b and Table 1) by volumetric recharge rates expressed in cubic meters per day (m^3/day). Recharge rate is estimated using the equation

$$R = \frac{\Delta h \times \eta \times A}{t}$$

where R is recharge rate (m^3/day); Δh is change in water level during irrigation event (m), derived using the hydrograph separation method (Fetter, 1988); η is porosity (as a unitless decimal, estimated at 25 percent (0.25); A is irrigated area (ha); and t is the duration of the irrigation event (days). The values for recharge reported in Table 1 were obtained by averaging recharge estimates based on water level measurements from all of the wells in a given irrigation plot. Complete water level and Assert concentration datasets can be found in Miller *et al.* (2002).

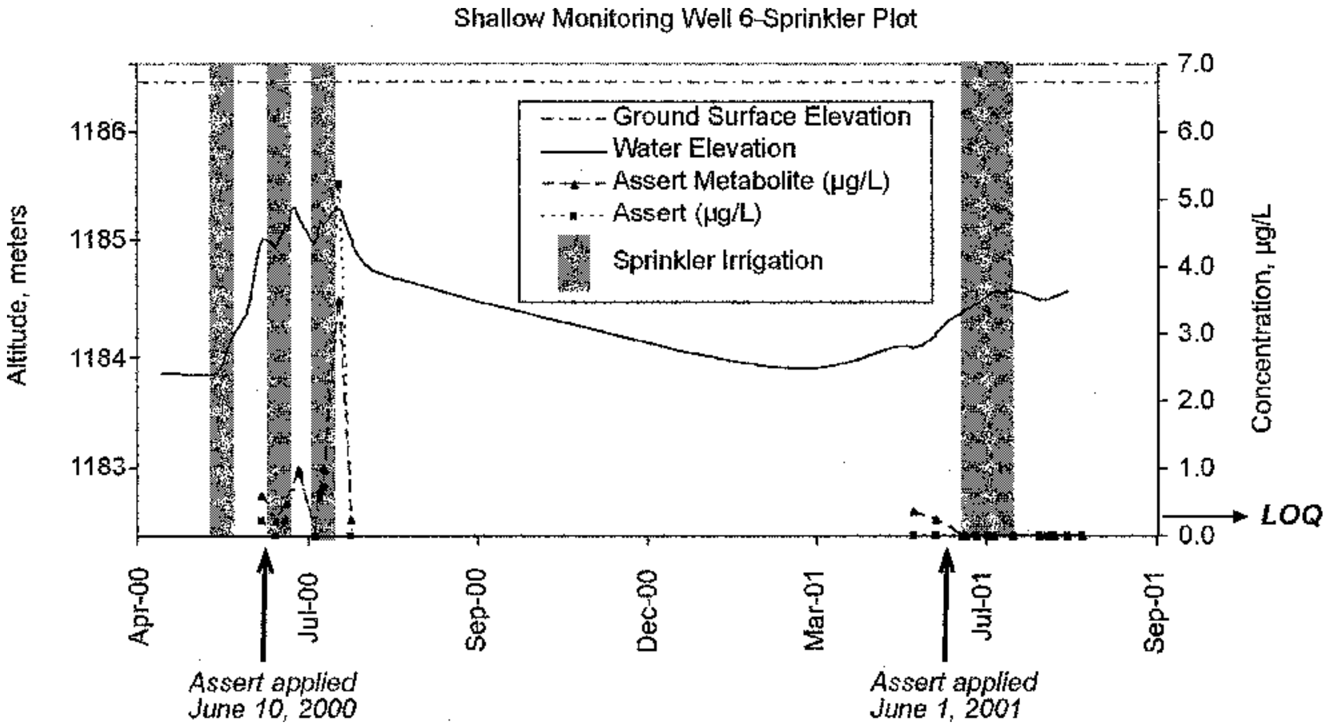
The duration of flood, wheel line, and center pivot irrigation events were 3, 12, and 18 days, respectively. Accordingly, the values for duration of recharge event as used in rate calculations (Table 1) were three days for flood events in 2000 and 2001, 12 days for wheel line sprinkling in 2000, and 18 days for center pivot irrigation in 2001. The analytical LOQ for Assert is 0.2 µg/L.

RESULTS AND DISCUSSION

Ground Water

Assert and metabolite concentrations in ground water across the entire study field were very low, ranging from < 0.2 µg/L to 10.0 µg/L (Table 2). Analysis of data reliability (Horwitz, 1982) suggests that because the concentrations were so low the data should not be used for trend analysis. Accordingly, all mass flux calculations are based only on maxima and minima. Background levels of Assert in ground water samples collected prior to the June 10, 2000, application of Assert to the field ranged from no-detection to the LOQ of 0.2 µg/L. Background levels of the Assert metabolite range from no-detection to 0.6 µg/L. Prior to June of 2000, Assert had not been applied to this field in the two years between spring 1997 and June 2000.

Assert concentrations in ground water samples collected on May 31, 2001, prior to that year's application were higher than those of June 10, 2000, ranging from the LOQ to 1.6 µg/L. The acid metabolite is generally found in higher concentrations than the Assert parent compound (Table 2). This may be explained by the rapid hydrolysis of the parent compound. Assert and metabolite concentrations are usually higher in shallow wells than in deep wells. The highest concentration of metabolite, 3.5 µg/L in 2000, was found in a shallow well in the sprinkler plot (Miller *et al.,* 2002).



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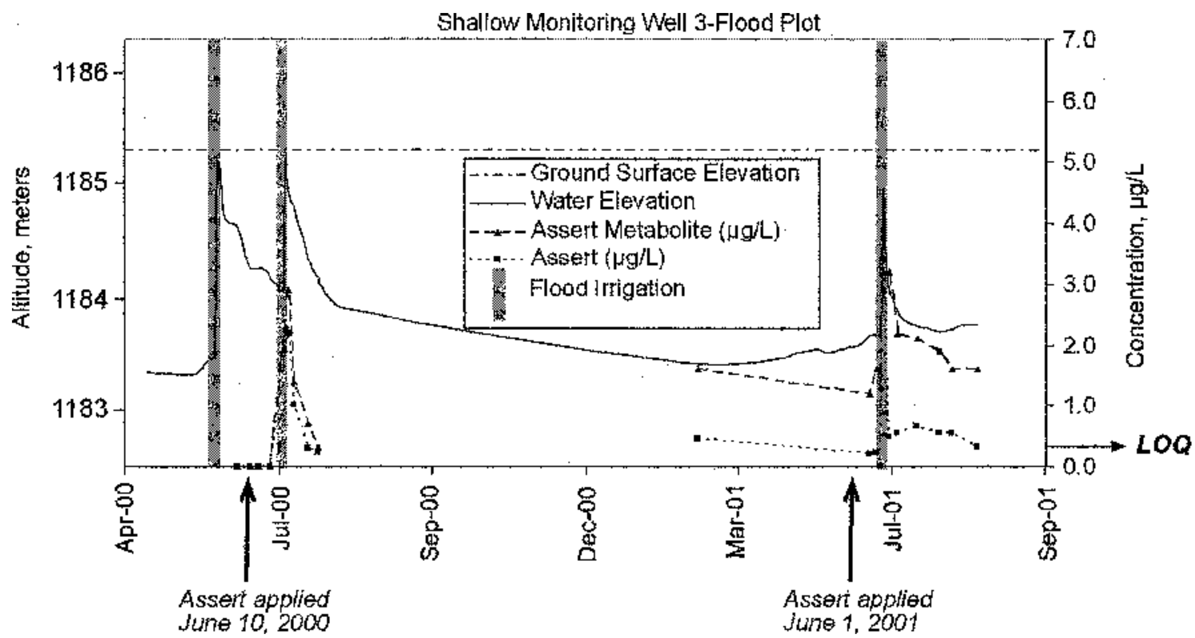

(b)

Figure 3. Hydrograph of (a) Monitoring Well 6 in the Sprinkler Plot; and (b) Monitoring Well 3 in the Flood Plot.

In 2001, the highest concentration of metabolite (10.0 µg/L) was found in a shallow well on the flood plot (Miller *et al.,* 2002). At the beginning of the growing season in 2000, higher concentrations of Assert were found in ground water beneath the sprinkler plot than beneath the flood plot. Concentration breakthrough in the shallow wells occurred about 30 days after application in the flood irrigated plot (2000 and 2001) and about 40 days after application (2000) in the wheel line sprinkler irrigated plot (Figures 3a and

TABLE 1. Average Values for Ground Water Recharge and Assert Metabolite Mass Flux Under Three Irrigation Methods.

Year	Irrigation Method	Duration of Irrigation Event (days)	Area Irrigated (hectares)	Recharge (cm)	Recharge Rate (m^3/day)	Metabolite Flux (grams/day)
2000	Flood	3	15	30	15,500	7.3
2001	Flood	3	3	34	3,540	8.3
2000	Wheel Line	12	45	9	3,170	5.1
2001	Center Pivot	18	57	3	854	0.0

TABLE 2. Ground Water Maximum, Minimum, and Mean Values of Assert and Metabolite in Samples from All Shallow Wells.

	Number of Samples*	Maximum Assert (µg/L)	Minimum Assert (µg/L)	Mean Assert (µg/L)	Maximum Metabolite (µg/L)	Minimum Metabolite (µg/L)	Mean Metabolite (µg/L)
Flood	279	8.3	0.0	0.2	10.0	0.0	0.6
Wheel Line	69	5.2	0.0	0.2	3.5	0.0	0.3
Center Pivot	107	0.0	0.0	0.0	0.0	0.0	0.0

*Values of 0.0 are equivalent to reported values of "no-detect."

3b). Breakthrough did not occur in 2001 in ground water beneath the center pivot sprinkled plot. Concentrations in the deep wells were generally lower, ranging from less than 0.2 to 0.5 µg/L.

The low density of Assert (0.3 g/mL at 20˚C) (WSSA, 1994) probably accounts for its preferential distribution in the upper portion of the water column. Additionally, the upper portion of the aquifer contains proportionately more clay that may function as a temporary reservoir for Assert. Other investigators have noted that in clay soils that are subject to wet/dry cycles similar to conditions on the Greenfields Bench, herbicides similar to Assert will diffuse into the clay micropores about four times faster than they diffuse out of the micropores after rewetting has occurred (Bryan Gentsch, BASF, March 1, 2002, personal communication).

Soil

In soils with an alkaline pH such as those found on the Greenfields Bench, Assert is expected to be weakly adsorbed. When these conditions occur, the pesticide is very soluble and will be highly mobile in soil unless a chemical complex is formed (Wauchope *et al.,* 1992) or unless binding occurs in soil clay and/or organic matter.

A summary of Assert concentrations in soil is in Miller *et al.* (2002). In general, concentrations of Assert and metabolite in soil were higher in 2001 than in 2000 (Table 3). Two years elapsed between the application in spring 1997 and soil sampling on March 8, 2000. But only nine months elapsed between Assert application on June 10, 2000, and soil sampling on March 3, 2001, leaving higher residual metabolite concentrations in soil in 2001 because of less time for transport or degradation of the metabolite than in the 2000 soil sample.

Even though Assert had not been applied to the field plots for two years prior to the initial soil sampling on March 8, 2000, as much as 120 mg/kg of the parent compound was still measured in soil samples. The extreme persistence of Assert in soil is problematic, and its presence after two years is inconsistent with the published soil half-life estimate of 45 days (Wauchope *et al.,* 1992). The soil appears to be serving as a "reservoir" for Assert, allowing the chemical to desorb or degrade. While the factors influencing the rate of desorption/degradation probably include mineralogy, pH, and micropore size, microbial degradation has been cited as the primary mode of chemical decomposition (WSSA, 1994). Evaluating agricultural practices with an eye towards optimizing soil health and microbial ecology will maximize microbial degradation of Assert and possibly reduce persistence in soil.

TABLE 3. Assert Concentrations in Soil.

		Flood Plot		Sprinkler Plot	
Date	Event	Assert (µg/kg)	Metabolite (µg/kg)	Assert (µg/kg)	Metabolite (µg/kg)
March 8, 2000	Pre-Assert Application	70	5	120	8
March 3, 2001	Pre-Assert Application	110	39	230	37

Ground Water Recharge and Mass Flux

Concentration data for the metabolite of Assert are statistically more reliable than data for the parent compound because the metabolite concentrations are significantly higher than those of the parent compound. Because of its increased data reliability, the discussion of herbicide mass flux is limited to an analysis of the metabolite of Assert.

The decrease in flood irrigated area from 15 ha in 2000 to 3 ha in 2001 (Table 1) resulted in a proportional decrease in recharge rates from 15,500 m^3/day in 2000 to 3,540 m^3/day in 2001. In contrast, metabolite flux increased from 7.3 g/day in 2000 to 8.3 g/day in 2001. The increased flux is probably attributable to less dilution in 2001 than in 2000. The higher metabolite flux in ground water at the flood plot in 2001 is also consistent with the higher Assert soil residuals in 2001 (Table 3). Recharge rates for ground water under the flood plot in 2001 (3,540 m^3/day) and the wheel line plot in 2000 (3,170 m^3/day) are similar. But at 8.3 g/day, the metabolite flux for the flood plot in 2001 is significantly higher than the 5.1 g/day flux rate for the wheel line plot. This difference may be caused by the additional transport mechanism that is often available during flood irrigation, in which an influx of Assert to ground water occurs with the establishment of a hydraulic connection between rising ground water and saturated, Assert laden soil. A comparison of recharge and metabolite flux between ground water under the wheel line sprinkler in 2000 and the center pivot in 2001 (854 m^3/day) shows decreased recharge in 2001 resulting in zero metabolite flux (Table 1).

In summary, flood irrigation provides about 30 cm of recharge (Table 1) that transports Assert and its metabolite to ground water, but recharge rates higher than 15,500 m^3/day may reduce concentrations through dilution. The 9 cm of recharge (Table 1) provided by the wheel line sprinkler is also sufficient to transport Assert through the soil into ground water but is insufficient to significantly reduce concentrations through dilution. It appears that the center pivot does not provide sufficient hydraulic loading to transport Assert from the soil downward into ground water.

CONCLUSIONS

Sprinkler irrigation of either type results in lower Assert metabolite flux to ground water than flood irrigation. Wheel line irrigation provides 9 cm of recharge, about 70 percent less than the 30 cm of recharge provided by flood irrigation. Center pivot irrigation provides 3 cm of recharge, about 90 percent less than flood irrigation. The variables that cause changes in the recharge and mass flux attributable to any one irrigation method include: (1) irrigation technique, such as flood versus wheel line versus centerpivot; (2) changes in the number of hectares that are irrigated by a given method in a given year; (3) differences in duration of irrigation event; and (4) differences in Assert concentrations in ground water under each irrigation method.

Public health risks are mitigated by the low toxicity and low concentrations of Assert and metabolite in drinking water. However, if a more toxic, but equally persistent and leachable, herbicide were to be used on the Greenfields Bench, concentrations in drinking water could be high enough to raise public health concerns and trigger regulatory action. The use of only center pivot irrigation may reduce ground water contamination, but the problem of herbicide persistence in soil and reduced aquifer recharge would still remain. Soils on the Greenfields Bench appear to function as a “reservoir” of Assert and its acid metabolite, allowing the chemical to desorb or degrade at a rate that varies with environmental conditions that remain unclear but that may include pH, organic matter, clay type, clay amount, micropore size, and microbial activity. Since soil provides a long term (> 2 years) source of Assert, evaluating all agricultural practices to optimize soil microbial activity will maximize microbial degradation of Assert and may reduce persistence in soil.

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