

GW10B: GROUND WATER PROTECTION LIST MONITORING FOR ORYZALIN

Kelly Aguirre
Scientific Aid

Murray Clayton
Staff Environmental Scientist

ABSTRACT

Oryzalin, an active ingredient on the Ground Water Protection List (GWPL), was selected for well monitoring because of its threat to ground water as predicted by a prioritization scheme that is based on use intensity and modeling results simulating movement of pesticides to ground water. Well selection was focused on the heaviest oryzalin use in sections of land that were considered vulnerable to offsite movement of pesticide residues. From December 2010 to April 2011, the Department of Pesticide Regulation (DPR) sampled 41 wells in Fresno, Kern, Madera, Merced, San Joaquin, and Tulare counties for oryzalin and several other herbicides previously found in California ground water by DPR (hexazinone, tebuthiuron, simazine, bromacil, prometon, atrazine, norflurazon, and diuron). DPR did not detect oryzalin in the sampled wells, yet several of the other monitored pesticides were detected in 18 wells. Oryzalin use in most of the sections containing these 18 wells and in the sections containing the remaining 23 wells was substantially heavier than the use of the pesticides that were detected. Furthermore, compared to the detected pesticides, there were no unique oryzalin use patterns that could account for its lack of detection in ground water. This suggested that it was oryzalin's physical/chemical properties preventing its movement to ground water. Compared to the detected pesticides oryzalin has a lower aqueous solubility, higher potential for soil adsorption, and faster dissipation in soil under both aerobic and anaerobic conditions. From the results of this current study and the lack of oryzalin detections in previous ground water monitoring studies conducted by DPR, it is unlikely that oryzalin is a threat to California ground water under its current labeled use directions and use patterns.

The GWPL prioritization scheme overestimated oryzalin's threat to ground water, most likely from over-weighting its use intensity component, which for oryzalin has historically been relatively high. Partitioning of greater weighting to the scheme's modeling component, which relies largely on the chemical's physical/chemical properties would have reduced oryzalin's predicted threat to ground water. Further ground water monitoring of other pesticides and investigations into reweighting of the various ranking components in the prioritization scheme will yield more insight into improving the scheme's predictive capabilities.

INTRODUCTION

DPR is required to monitor ground water for pesticides on the GWPL to determine if these pesticides have migrated to ground water as a result of legal agricultural use. In California, a pesticide is placed on the GWPL and subject to monitoring if it has specific labeled uses and it exceeds threshold values, termed specific numerical values (SNVs), for certain combinations of

physical/chemical properties (Johnson 1991, 1989 and 1988). Pesticides on the GWPL are prioritized for monitoring based on use intensity, physical/chemical properties and current registration status. Registered pesticides with heavy and increasing use with a higher potential to move to ground water, based on computer simulations with the LEACHM pesticide fate and transportation model (Hutson and Wagenet, 1992) receive a higher prioritization because they present a greater potential threat to ground water.

The herbicide oryzalin exceeds the mobility SNV for organic carbon-normalized soil adsorption coefficient (Koc) and the persistence SNVs for anaerobic soil metabolism and hydrolysis half-lives (Table 1), and has soil-applied uses qualifying it for placement on the GWPL. DPR selected oryzalin for monitoring due to its perceived high potential to migrate to ground water. The current GWPL prioritization scheme has oryzalin ranked as the fourth highest threat to California ground water. Despite being less mobile and persistent than pesticides currently regulated as ground water contaminants (Table 1), oryzalin's potential threat to California ground water remains uncertain considering its exceedance of the SNV's and highly ranked threat by the GWPL prioritization scheme.

Between 1993 and 2006, the U.S. Geological Survey (USGS) detected oryzalin in 12 ground water wells in several states other than California at concentrations ranging from 0.012 to 0.57 ug/L (USGS, National Water Quality Assessment Data Warehouse). Land use where ten of the oryzalin detections occurred was classified as urban (residential/commercial) or mixed. Land use overlaying the remaining two detections was agricultural and occurred in Georgia in shallow wells less than 65 feet deep. Oryzalin concentration in these wells was 0.016 and 0.0285 ug/L; both were remarked as estimates because of their low levels. To date there has been no detection of oryzalin in California ground water by other local, state or national agencies.

However, use of oryzalin in California has been high. From 1998, use both statewide and in ground water protection areas (GWPA) decline rapidly following a period of high use because of oryzalin production-related issues. After 2001, use of oryzalin steadily increased, returning to its previously high levels (Figures 1 and 2). Sites in California which account for the most use of oryzalin include almonds (23 percent [%] of total use), wine grapes (17%), other grapes (12%), pistachios (10%), and rights of way (10%).

The historical and extensive use of oryzalin in California has initiated a number of monitoring studies. DPR cumulatively sampled 130 wells for oryzalin in 1993, 1998, and 2007 without detecting any of its residues. However, many of these wells were in sections specifically targeted for the pesticide napropamide, which was a constituent in the analytical screen for oryzalin. With little overlapping use between these pesticides potential detections of oryzalin were not expected in the wells targeted for napropamide. In 1993 only two wells were sampled for oryzalin at a reporting limit (RL) of 0.05 ug/L. In 1998, samples from 54 wells were tested for oryzalin, also at a RL of 0.05 ug/L, but only 25 of these wells were in sections specifically targeted for oryzalin; 20 wells were in sections targeted for napropamide and the remaining nine wells were in sections target for both oryzalin and napropamide (Weaver, 1999). In 2007, 74 wells were sampled for oryzalin at a RL of 0.05 ug/L. Fifty-one wells were in sections specifically target for oryzalin and the remaining 23 wells were in sections targeted for napropamide (Fossen, 2008). In

that study, well sampling was restricted to the most vulnerable GWPA's with an average depth to ground water of less than 30 feet. Consequently, well sampling locations were highly clustered being limited to either one township or two contiguous townships in each county. Merced was the exception with wells in four townships sampled; however, all four townships were contiguous (Fossen, 2008). High oryzalin use did factor in the selection of land sections for ground water monitoring. Median cumulative use in sections targeted for oryzalin monitoring in 1998 and 2007 was 1,328 pounds (lbs) (use from 1991-1995) and 2,078 lbs (use from 1992-2005), respectively.

From February to April 2010, DPR sampled approximately one-third, or 23 wells for oryzalin from DPR's domestic well monitoring network located in Fresno and Tulare counties. DPR established this network in 1999 to assess the impact of pesticide use restrictions on the detected concentrations of regulated pesticides and their degradates (Garretson, 1999). Sampling these wells for oryzalin provided an opportunity to further evaluate its potential to contaminate ground water in areas with wells known to have been impacted by pesticides. The 23 wells selected for sampling were in sections with the highest cumulative use of oryzalin from 1990 to 2010. While one section had exceptional high oryzalin use of 7,328 lbs (Table 2), the median use in these network sections was only 1,365 lbs. Oryzalin was not detected in the network wells (Garretson, 2012).

State-wide use of oryzalin has remained high through 2010. This current study was designed to monitor for oryzalin in the highest use sections in GWPA's covering a broad area of the Central Valley to more fully resolve its potential threat to ground water in California. The study also provided an opportunity to assess the predictive capabilities of the GWPL prioritization scheme.

MATERIALS AND METHODS

This current monitoring study had the goal of sampling 40 wells in unique sections of high oryzalin use, particularly in those sections where cumulative use was greater than the cumulative use in most of the sections sampled by DPR in the 1998, 2007, and 2010 oryzalin studies. This study focused also on sampling for oryzalin in GWPA's because they are considered vulnerable to ground water contamination. GWPA's have a shallow ground water table of 70 feet or less and either coarse textured soils prone to leaching or semi-impermeable soil layers limiting water infiltration that increases the potential for surface run-off to leaching vulnerable soils, either inadvertently or through engineered sites or structures. A limited number of sections without soils data or depth-to-ground water data have been designated GWPA's because pesticides residues have been found in ground water in these sections. DPR's pesticide use reporting database was used to identify 40 high priority sections for sampling with the highest oryzalin use in GWPA's. An additional 40 alternate sections, also in GWPA's with substantial oryzalin use were identified for potential sampling to substitute for any high priority sections that could not be sampled.

Within targeted sections, wells were selected with close proximity to almonds, wine grapes, other grapes, and pistachios – crops with the heaviest reported use of oryzalin. DPR selected domestic wells for sampling with the goal of sampling one well per section according to standard operating procedure (SOP) FSWA006.01 (Nordmark and Pinera-Pasquino, 2008). If a suitable

well was not available in the target section, a well within approximately 0.2 miles of the section could be sampled as long as it was at least one mile away from a previously sampled well. Samples were collected using the methods described in SOP FSWA001.01 (Nordmark and Herrig, 2011).

The California Department of Food and Agriculture's (CDFA's) Center for Analytical Chemistry analyzed two primary samples from each well, one for oryzalin and one collectively for several pesticides previously found in ground water by DPR: hexazinone, tebuthiuron, simazine, bromacil, prometon, atrazine, norflurazon, diuron, and some of their degradates (CDFA, 2009; and CDFA, 2010). Samples containing a known amount of analyte, disguised as actual samples (blind spikes), were prepared and analyzed in accordance with SOP QAQC001.00 (Segawa, 1995). Samples containing deionized water (field blanks) were collected at the same time as the well water samples in accordance with SOP QAQC011.02 (Richardson, 2011). They were tested as a quality control measure for potential contamination resulting from sample collection procedures when corresponding well water samples tested positive for one or more analytes. Samples from sites coded 10-08 thru 10-11 and 20-04 thru 20-06 deviated from this SOP in that purified water was used in lieu of deionized water. The RL for all analytes was 0.05 ug/L. The RL is the lowest concentration that can be reliably detected and is set by the testing laboratory for each compound. The oryzalin method and the method used for the collective group of pesticides previously found in ground water and monitored for in this current study have been determined to be unequivocal (Aggarwal, 2011; and Fattah, 2008).

RESULTS

Analysis of pesticide use reporting for oryzalin between 1995 and 2007 identified 40 sections inside GWPAs with the highest cumulative use of oryzalin – more recent use up to 2010 was not included in cumulative totals because this use is likely too recent to have potentially impact ground water. Cumulative use in all of these sections exceeded 2,700 lbs, surpassing the median cumulative use in sections containing wells sampled in the 1998 and 2007 oryzalin studies and in DPR's well network. Cumulative oryzalin use in the 40 alternate sections that also were located inside GWPAs ranged from 2,060 to 2,700 lbs between the years 1995 and 2007. Table 3 lists the sections sampled and cumulative applications of oryzalin. Of the collective group of pesticides that also were monitored for in this current study only cumulative use for diuron, simazine and norflurazon are listed in Table 3 because only these pesticides were detected in this study.

From December 2010 to April 2011, 40 wells plus one additional well were sampled in unique sections in Fresno, Kern, Madera, Merced, San Joaquin, and Tulare counties. Oryzalin was not detected in any wells (Table 3). However, 18 wells located in Fresno, Madera, San Joaquin, and Tulare counties tested positive for one or more other pesticides and/or degradates. Diuron was detected in four wells at concentrations ranging from 0.066 to 0.188 parts per billion (ppb). Norflurazon was found in four wells at concentrations ranging from 0.064 to 0.211 ppb. Simazine was detected in three wells at concentrations ranging from 0.078 to 0.095 ppb. Deethyl simazine or deisopropyl atrazine (both chemically identical and abbreviated as ACET) – primary degradates of simazine or atrazine – were found in ten wells at concentrations ranging from 0.075 to

0.934 ppb. Diamino chlorotriazine (DACT) – secondary degradate of simazine or atrazine – was found in 17 wells at concentrations ranging from 0.05 to 2.17 ppb. Desmethyl norflurazon (DSMN) – primary degradate of norflurazon – was detected in ten wells ranging in concentration from 0.076 to 1.30 ppb (Table 3).

DISCUSSION

Potential for Oryzalin Movement to Ground Water

Oryzalin was not detected in ground water in this current study despite being targeted for in vulnerable areas with its highest reported use. Vulnerable sections were identified by their classification as a GWPA. All but 2 of the 41 wells sampled in this study were in sections classified as GWPAs because they had soil type and depth-to-ground water characteristics associated with offsite movement of pesticides to ground water as determined by the CALVUL model (Troiano et al., 2000). The detections of diuron, norflurazon, simazine, or their degradates in 18 wells in unique sections confirmed that pathways for residue movement to ground water existed in almost half of the sections sampled. Although oryzalin residues were not detected in these 18 wells cumulative use of oryzalin exceeded the use of the detected pesticides in 12 of the 18 sections (Table 3). For all 41 sections in which wells were sampled median cumulative use of oryzalin was 2,805 lbs. For diuron, norflurazon, and simazine median cumulative use was 104, 309, and 1,190 lbs, respectively. The lack of oryzalin detections was therefore not likely due to insufficient use intensity. Nor was it likely due to use practices as oryzalin, diuron, norflurazon, and simazine are all pre-emergent soil applied herbicides to bare soil used primarily on nut, fruit, and vine crops, and rights-of-way (CDPR, 2011b). Diuron and norflurazon also have prominent use in alfalfa (CDPR, 2011b).

The most likely mechanism preventing oryzalin's movement to ground water is its chemical/physical properties. Despite exceeding several SNVs, oryzalin's mobility and persistence properties are less severe than those for diuron, norflurazon and simazine, and other pesticides found in ground water by DPR (Table 1). Oryzalin has been reported as persistent but not mobile under field conditions (U.S. Environmental Protection Agency, 1994). Aqueous solubility for oryzalin is 2.6 parts per million, which does not exceed the mobility SNV for solubility and is considerably lower than those of all the pesticides previously detected in ground water and regulated by DPR (Table 1). However, oryzalin's Koc value of 807 cm³/g fails this mobility SNV threshold but is still substantially higher with a greater soil adsorptive potential than those Koc values for DPR's regulated pesticides (Table 1). Diuron, a pesticide that is regulated as a ground water contaminant in California has been shown to be more mobile than oryzalin in lab and field studies that compared their leaching characteristics (Landry, 2004 and 2006). In both studies, the movement of diuron in the soil was twice as deep compared to the movement of oryzalin. This was almost certainly due to oryzalin's lower solubility and higher capacity for soil adsorption compared to diuron. Results from these studies are consistent with findings from this oryzalin monitoring study suggesting that the physical/chemical properties of oryzalin are not conducive for its movement to ground water, even in areas of California that are vulnerable to off-site movement of pesticide residues.

Potential for Oryzalin Degradate Movement to Ground Water

Degradation products of oryzalin were not included in the analytical method for this current study because certified analytical standards were not available. Krieger et.al. (1998) investigated dissipation and sorption patterns for oryzalin and seven of its degradation products in four soil types under laboratory aerobic conditions. The authors found that during a six month incubation period no single degradation product exceeded 10% of the original oryzalin application rate in any of the four soil types. They also noted that four of the seven degradates originated directly from the oryzalin molecule, which may explain the low recovery of any single degradation product relative to the parent application rate. These data indicate it would be unlikely that oryzalin degradation products would be detected in ground water at our current RL.

GWPL Prioritization Scheme

The GWPL prioritization scheme ranked oryzalin as having a high potential to impact ground water relative to other listed pesticides. While this ranking for oryzalin exceeded rankings for most of the regulated pesticides on the GWPL, it was oryzalin's heavy state-wide use and its disproportionately heavier use in GWAs that significantly elevated its relative ranking. The integration of the modeling- and use-based ranking components positioned oryzalin as the fourth highest predicted threat to ground water, only to be exceeded by diuron, norflurazon, and metolachlor or S-metolachlor (a stereoisomer of metolachlor). Diuron, norflurazon, and desmethyl norflurazon (degradate of norflurazon) have been found in ground water by DPR in numerous locations (CDPR, 2012) including 12 sites in this study. Metolachlor has been found by the USGS in two wells in California (CDPR, 2011c) and two of its degradation products, ethane sulfonic acid and oxanilic acid have been found by DPR in 64 and 23 wells, respectively (CDPR, 2012). Simazine, hexazinone and bromacil are ranked immediately below oryzalin on the GWPL prioritization scheme. These pesticides also have been detected by DPR in numerous locations throughout California (CDPR, 2012). Currently, diuron, norflurazon and bromacil are regulated within GWAs. Hexazinone is currently not regulated because it has not been detected at concentrations that threaten public health (Pritchard, 2011). Metolachlor and S-metolachlor are currently under review by DPR.

The absence of detections of oryzalin in this current study and in previous ground water monitoring studies contrasts with both its elevated threat to ground water predicted by the prioritization scheme and its comparable ranking among other pesticides found in California ground water. Contrasting further with the scheme's prediction capabilities are the recent detections of tebuthiuron in 12 wells (Dias, 2011) and the mostly historical yet continued detections of prometon throughout California (CDPR, 2012) despite its negligible use over the past 20 years (CDPR, 2011b). The scheme underestimated the threat to ground water for these two pesticides, yet they have the highest predicted threats to ground water when based solely on the prioritization scheme's modeling component. This component to the prioritization scheme simulates potential movement of pesticides to ground water and is based only on their physical/chemical properties and application rate. Tebuthiuron and prometon's overall deemphasized risk to ground water by the prioritization scheme resulted from their relatively minor use in California compared to other pesticides that have been detected in ground water. These facts, including results from this monitoring study suggest that the pesticide use

components in the scheme's ranking procedure might be less important at associating threats to ground water than the scheme's modeling component, or the physical/chemical properties of pesticides. Further ground water monitoring and investigations into reweighting of the various ranking components in the prioritization scheme will yield more insight into potentially optimizing the scheme's prediction capabilities.

CONCLUSIONS

The physical/chemical properties of oryzalin indicate on the whole it is less mobile and persistent in soil compared to pesticides that have been detected in California ground water and currently regulated by DPR. This would explain the lack of detections of oryzalin in this current monitoring study and in previous monitoring studies, and the unlikelihood that oryzalin will impact California ground water under its current labeled use directions and rates of application, and intensity of use in vulnerable areas.

Data from this current study also suggest that oryzalin's highly elevated threat to California ground water, determined from its ranking on the GWPL prioritization scheme, has been overestimated. This is likely resulting from the scheme's weighting on oryzalin's historical use, which has been substantial compared to most of those pesticides found in ground water and currently regulated by DPR. Numerous ground water detections of several pesticides which exceed the SNVs for mobility and persistence, yet have modest state-wide use indicates that movement of pesticides to ground water might be more associated with physical/chemical properties than with use intensity.

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FIGURES

Figure 1. Oryzalin Use in California (CDPR, 2011b).

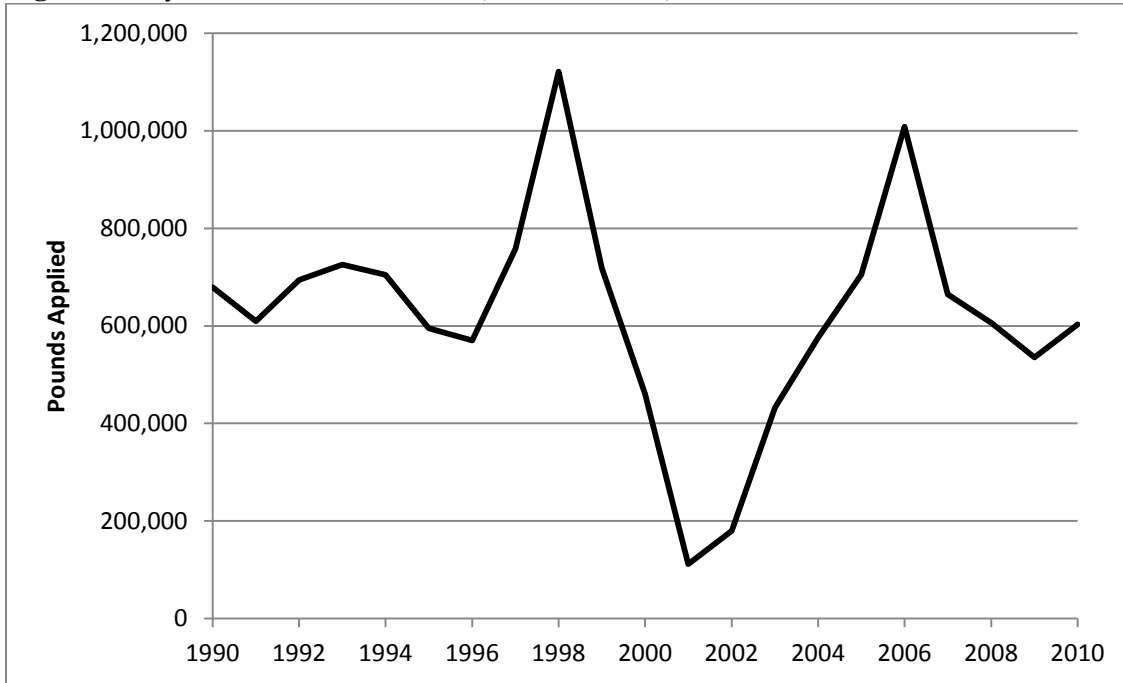
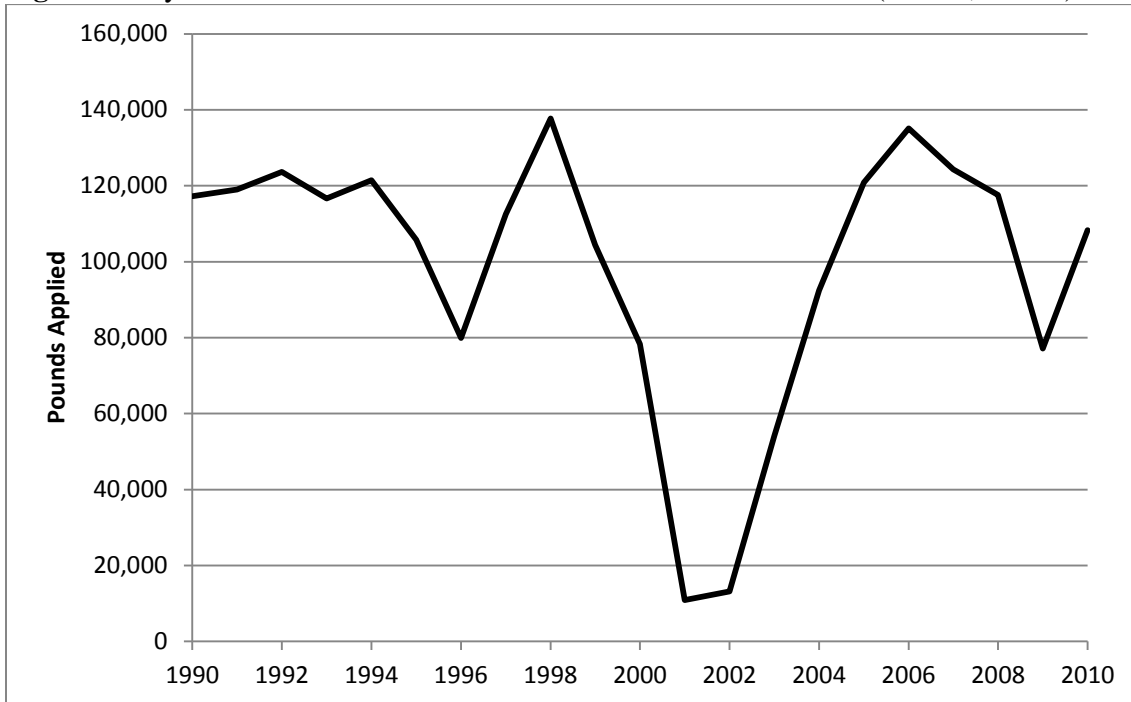


Figure 2. Oryzalin Use in California Groundwater Protection Areas (CDPR, 2011b).



TABLES

Table 1. Specific numerical values (SNV; 3, California Code of Regulations section 6804) and physical-chemical properties for oryzalin, atrazine, bromacil, diuron, norflurazon, prometon, and simazine (CDPR, 2011a).

	Mobility		Persistence		
	Water solubility (parts per million)	Koc (cm ³ /g)	Aerobic soil metabolism (days)	Anaerobic soil metabolism (days)	Hydrolysis (days)
SNV	>3	<1900	>610	>9	>14
Oryzalin	2.60	807	63.3	10	stable
Atrazine	32.5	92.9	146	159	stable
Bromacil	929	17.3	347	72.5	stable
Diuron	36.4	499	372	995	1290
Norflurazon	33.7	617	172	348	2650
Prometon	718	124	459	61	1130
Simazine	6.15	340	110	70.8	stable

Table 2. Well monitoring network sections sampled for oryzalin. Pounds of oryzalin applied in each section are cumulative from 1990-2010 (CDPR, 2011b).

County	Section	GPWA	Pounds of oryzalin applied
Fresno	10M14S21E25	R	2,584
	10M14S22E12	R	2,935
	10M14S22E31	L	1,937
	10M14S22E33	L	1,365
	10M14S23E32	L	2,686
	10M14S23E33	L	7,328
	10M14S23E34	L	1,072
	10M14S23E35	R	1,522
	10M15S21E09	L	959
	10M15S22E15	L	1,846
	10M15S22E16	L	1,306
	10M15S23E12	R	1,105
	10M15S24E14	R	880
	10M16S21E34	L	2,312
	Tulare	54M16S23E01	L/R
54M16S24E14		R	2,788
54M17S26E26		R	1,454
54M18S27E29		R	947
54M19S26E01		R	919
54M20S26E03		R	2,309
54M20S26E24		R	1,344
54M20S27E20		R	1,207
54M20S27E31		R	822

L = Leaching GWPA

R = Runoff GWPA

San Joaquin	01S/07E-07	39-02	L	4,031	63	96	451	0.000	0.000	0.000	0.000	0.000	0.000	0.000
San Joaquin	02S/09E-07	39-03	L	3,175		563	564	0.000	0.000	0.000	0.000	0.175	0.078	0.490
San Joaquin	02S/08E-02	39-04	L	4,119	642	717	2,108	0.000	0.000	0.000	0.000	0.000	0.000	0.000
San Joaquin	02S/08E-07	39-05	L	3,964	34	129	973	0.000	0.000	0.000	0.000	0.000	0.000	0.000
San Joaquin	02S/08E-09	39-06	L	5,357		337	1,391	0.000	0.000	0.000	0.000	0.000	0.000	0.175
San Joaquin	02S/08E-06	39-07	L	1,321 (3,292) ^c	24	301	555	0.000	0.000	0.000	0.000	0.000	0.000	0.000
San Joaquin	01S/08E-30	39-08	R	11,350		331	2,640	0.000	0.000	0.074	0.000	0.076	0.306	0.267
San Joaquin	02S/08E-03	39-09	L	2,338	112	433	3,039	0.000	0.000	0.000	0.000	0.000	0.000	0.000
San Joaquin	02S/09E-14	39-11	L	2,955	20	569	1,190	0.000	0.000	0.000	0.000	0.216	0.000	0.127
San Joaquin	02S/09E-16	39-12	L	1,869 (4,551) ^c	1,154	657	2,011	0.000	0.000	0.000	0.000	0.233	0.090	0.349
San Joaquin	02S/09E-09	39-13	L	3,035	116	590	1,249	0.000	0.000	0.211	0.000	1.300	0.000	0.251
San Joaquin	02S/09E-06	39-14	L	2,785	203	1,338	2,030	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Tulare	16S/23E-09	54-01	L	3,398	44	138	1,914	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Tulare	17S/23E-02	54-02	R	3,638		365	1,869	0.000	0.076	0.064	0.078	0.388	0.232	0.457
Tulare	17S/23E-22	54-03	R	397 (2,684) ^c	1,342	52	1,361	0.000	0.066	0.000	0.000	0.000	0.121	0.448
Tulare	21S/26E-11	54-04	R	3,615	811	1,480	837	0.000	0.000	0.000	0.000	0.000	0.000	0.154
Tulare	18S/26E-32	54-05	R	38 (2,064) ^c	317	24	68	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Tulare	18S/27E-17	54-06	R	110 (2122) ^c	3,584	474	4,512	0.000	0.070	0.081	0.000	0.259	0.934	2.170

a. Sampled section was not a GWPA but was within 0.2 miles of a target section. Target section was a leaching GWPA.

b. Sampled section was classified as a leaching GWPA, but target section within 0.2 miles was classified as a runoff GWPA.

c. Sampled section was within 0.2 miles of targeted section. Target section use is given in parentheses.

LOC = Location code established during sampling

GWPA = Ground Water Protection Area

DSMN = Desmethylnorflurazon

ACET = Deisopropyl Atrazine

DACT = Diamino Chlorotriazine

R = Runoff GWPA

L = Leaching GWPA